Frequency-dependent conductivity of insulating Si:P and Si:As near the metal-insulator transition

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The microwave electrical conductivity $\sigma(\omega, N, T)$ of insulating Si:P has been measured for frequencies between 0.1 and 2.1 GHz in the density range $0.46 < N/N_c < 0.86$ ($N_c = 3.74 \times 10^{18}$ cm⁻³) and for temperatures between 1.5 and 4.2 K. For T < 2 K $\sigma(\omega, N, T)$ was found to be nearly independent of temperature, and showed a frequency dependence of the form $\sigma(\omega, N) \propto \omega^{s(N)}$ with s(N) increasing rapidly with increasing donor density from $s \sim 1$ to s > 2, and a very strong density dependence. These results are compared with similar measurements in Si:As and discussed in terms of pair photon-assisted hopping theories including and excluding electron-electron interactions. We also address the importance of extending these experiments into the dilution refrigerator temperature range.

I. INTRODUCTION

The process of electrical conduction in disordered systems near the metal-insulator transition (MIT) has been of considerable interest in recent years. The dc conductivity of uncompensated doped semiconductors, in particular, has been well documented on both the insulating 1^{-5} and the metallic 5^{-7} sides of the transition. It shows scaling behavior dependent on a localization length (correlation length on the metallic side) $\xi = \xi_0 |1 - N/N_c|^{-\nu}$. The temperature-dependent dc conductivity for $N < N_c$ has been observed to follow variable-range hopping (VRH) behavior $\sigma(T) = \sigma_0 \exp[-(T_0/T)^m]$ where *m* is predicted to be $\frac{1}{4}$ by Mott⁸ and $\frac{1}{2}$ by Efros and Shklovskii⁹ if one includes electron-electron interactions. $m = \frac{1}{4}$ has been measured in *n*-type Ga:As,¹⁰ in Ge:Sb,¹¹ Si:P,^{3,4} and Si:As.^{1,2,5} On the other hand, an exponent equal to $\frac{1}{2}$ has been seen in Ga:As (Ref. 12) and Ge:As.¹³ The ac conductivity of insulating doped semiconductors, however, is still not fully understood. Experiments have been mostly restricted to the low-frequency range ($v = \omega/2\pi < 1$ MHz) and the infrared regime. In the former case the ease of electrode placement on the sample's surface is counteracted by the presence of Schottky barriers which complicate data analysis if their impedance is comparable to the sample's. There have been few microwave measurements, which are difficult to perform and usually limited to the (fixed) resonant frequency of the cavity being used. The experimental information available is relatively small with respect to the theoretical work done and thus it is still not clear which of the proposed conduction processes, if any, can adequately explain the frequency, density, and temperature dependence of the conductivity $\sigma(\omega, N, T)$ of barely insulating doped semiconductors. The purpose of this paper is to report recent measurements¹⁴⁻¹⁶ of frequency-dependent conductivities in the microwave range between 0.1 and 2.1 GHz for barely insulating n-type Si samples and to compare the results with the various theoretical predictions for $\sigma(\omega, N, T)$. The low microwave range is useful for distinguishing between s < 1 and s > 1 frequency dependences of $\sigma(\omega, N, T \rightarrow 0) = [\sigma(\omega, N, T \rightarrow 0) \propto \omega^{s(N)}]$; most pair theories yield s < 1 for phonon-assisted hopping and s > 1 for photon-assisted hopping, as will be discussed below. We study the barely insulating regime for $0.46 < N/N_c < 0.86$ ($N_c = 3.74 \times 10^{18}$ cm⁻³) because earlier microwave work, to be reviewed below, has been done on more dilute samples.

In the dilute density range, low-frequency measure-ments $^{17-19}$ have established the sublinear frequency dependence of the conductivity $\sigma(\omega, T) \propto T \omega^s$ (s < 1). These results were obtained in the liquid-He temperature range and were explicable in terms of phonon-assisted pair-model hopping.²⁰ Low-frequency results have been reported recently by Paalanen et al.²¹ on a barely insulating $(N=0.96N_c)$ Si:P sample. Their measurements yielded an s < 1 frequency dependence that is finite as $T \rightarrow 0$ and shows a T^2 dependence at finite temperatures. Their results do not support the pair hopping mentioned above²⁰ and might be viewed as supporting a theory incorporating electron-electron interactions by Shklovskii and Efros²² for $\sigma(\omega, T=0$ K). An extensive study of ntype Si:As by Castner et al.²³ showed that the conductivity of a sample with $N = 0.87 N_c$ was independent of frequency up to 100 HZ, exhibited s < 1 for frequencies between 100 Hz and 100 kHz with a temperature dependence that becomes considerably stronger as T decreases below 295 mK. At frequencies above 100 kHz and temperatures less than 60 mK, a T-independent ω^2 law for $\sigma(\omega, T \rightarrow 0)$ in qualitative agreement with Mott's law was observed.

In the microwave range (100 MHz < v < 100 GHz) experiments have usually been made at fixed frequency, as mentioned above. Hess *et al.*²⁴ have made measurements (to be discussed further below) of Si:P at 400 MHz between 20 and 120 mK. At 9 GHz in dilute *p*-type Si $\sigma(\omega)$ was shown²⁵ to be proportional to 1/T and explained in terms of direct photon absorption of a pair. Recently Deri and Castner (DC) (Ref. 14) obtained a superlinear dependence for Si:As samples (concentration range $0.48N_c < N < 0.75N_c$) in the frequency range 100 MHz to

cy, density, and temperature ranges. Infrared absorption coefficient studies have been made²⁶⁻³⁰ in *n*-type Si with dopant density near the critical concentration. Si:P, the most studied MIT system was comprehensively studied^{29,30} in the infrared range as $N \rightarrow N_c$. These types of measurements show a superlinear frequency dependence^{26,29} near 10¹² Hz but the results are complicated by intradonor transitions among the donor 1s states.²⁹

II. THEORETICAL PREDICTIONS

Theories dealing with the ac conductivities of insulating doped semiconductors have been made both for T=0zero-phonon processes involving direct photon absorption and for phonon-assisted temperature-dependent pair-hopping processes.

Mott³¹ has obtained a T=0 ac conductivity of the form

$$\sigma(\omega) = \frac{\pi^2 e^2}{3\hbar} [N(E_F)]^2 (\hbar\omega)^2 \xi r_{\omega}^4 , \qquad (1)$$

where ξ is the localization length, $r_{\omega} = \xi \ln(2I_0/\hbar\omega)$, I_0 is the prefactor of the resonance integral between the states, and $N(E_F)$ is the density of states at the Fermi level. Equation (1), which we will refer to as Mott's ω^2 law, predicts a superlinear frequency-dependent conductivity with exponent of magnitude less than 2 which depends on the relative magnitude of I_0 and $\hbar\omega$.

Scaling theories of localization³²⁻³⁷ also predict $\sigma(\omega) \propto \omega^2$ but only for $\omega < \omega_c$ above which $\sigma(\omega) \propto \omega^{1/3}$. ω_c is a crossover frequency given by^{32,38} $\omega_c \simeq 1/$ $[\hbar N(E_F)\xi^3]$ which scales to zero as $N \rightarrow N_c$.

Shklovskii and Efros (SE) (Ref. 22) have corrected Mott's ω^2 law to include electron-electron interactions, obtaining

$$\sigma(\omega) = \frac{\pi^2 e^2}{3\hbar} [N(E_F)]^2(\hbar\omega) \xi r_{\omega}^4 \left[\hbar\omega + \frac{e^2}{\epsilon r_{\omega}} \right], \qquad (2)$$

where one recognizes the Coulomb interaction term of the pair within the parentheses [the SE (Ref. 22) theory only includes pair Coulomb forces and does not take into account long-range Coulomb interactions]. Because of the scaling behavior of ϵ and of the localization length ξ , the Coulomb interaction term scales to zero as N approaches N_c . The relative importance of the pair Coulomb interaction energy (which we shall call E_c with $E_c = e^2/\epsilon r_{\omega}$), as the dopant density changes, with respect to $\hbar\omega$ in Eq. (2), qualitatively predicts an exponent s(N)of the conductivity $\sigma(\omega) \propto \omega^{s(N)}$ that increases with N: If $E_c << \hbar\omega$ then Eq. (2) reduces to the Mott expression Eq. (1) and predicts 1 < s(N) < 2; if $E_c \gg \hbar\omega$ then Eq. (2) gives s(N) < 1. In the latter case the low-frequency regime of $\epsilon(\omega, N \rightarrow N_c)$ would diverge as $\omega \rightarrow 0$. Equation (2) was derived for the case of a small Coulomb gap $\Delta_g \ll e^2/\epsilon r_{\omega}$. In the opposite regime of a strong Coulomb gap $\Delta_g \gg e^2/\epsilon r_{\omega}$, SE obtain $\sigma(\omega)/\ln(2I_0/\hbar\omega)$. SE have extended these results to finite temperatures such that $kT \gg \hbar\omega$ and find $\sigma(\omega) \propto \omega^2 r_{\omega}^3/kT$. Other authors, ^{39,40} however, conclude that one obtains the *T*-independent ω^2 law as long as $kT \ll \langle V_0^2 \rangle^{1/2}$ with $\langle V_0^2 \rangle^{1/2}$ the width of energy distribution or a measure of the disorder.

Many authors have considered the relaxation or phonon-assisted hopping processes and the conductivity in this case assumes the form $\sigma(\omega, T)$ $\propto [N(E_F)]^2 \xi r_{\omega}^4 f(kT, \hbar \omega)$ where r_{ω} is now $(\xi/2) \ln(v_p/\omega)$ with v_p a phonon frequency. Austin-Mott²⁰ phononassisted hopping is obtained with $f(kT, \hbar \omega) = kT \hbar \omega$ unless Coulomb interactions are dominant (correlated hopping), $e^2/\epsilon r_{\omega} >> kT$, in which case the kT in the Austin-Mott result is replaced by $e^2/\epsilon r_{\omega}$. When $kT \ll 2I_0 r_{\omega}$, Bottger and Bryksin⁴⁰ have shown that the conductivity becomes

$$\sigma(\omega) = \frac{2\pi^5}{9} e^2 \xi [N(E_F)]^2 k T v_p (kT/2I_0) 2r_T^4$$

where $r_T = \xi/2 \ln(2I_0/kT)$. One thus obtains a frequency-independent, temperature-dependent conductivity. Castner and Deri¹⁵ have estimated that the cross-over from the Austin-Mott sublinear frequency dependence to frequency independent behavior can occur near 1 MHz at T=2 K. Finally, a prediction^{41,42} in the large-Coulomb-gap regime $\Delta_g \gg e^2/\epsilon r_{\omega}$ leads to the sublinear prediction $\sigma(\omega) \propto \epsilon' \omega / \ln(v_p/\omega)$ where ϵ' is the optical dielectric constant.

Following the discussion of Castner and Deri¹⁵ we draw these conclusions.

(1) Zero-phonon processes (except Ref. 22) yield superlinear frequency-dependent conductivities $[\sigma(\omega) \propto \omega^s, s > 1]$, while phonon-assisted processes (except Ref. 41) yield sublinear frequency dependences (s < 1).

(2) The zero-phonon (photon-assisted hopping) conductivity can exceed the phonon-assisted conductivity even for $\hbar\omega \ll kT$, contrary to predictions of simple hopping models based on impurity pairs.

(3) The absence of an observed temperature dependence does not conclusively rule out phonon-assisted hopping.

(4) The parameters I_0 and v_p have only been calculated in the dilute limit. There is no theoretical treatment of these quantities in the critical regime, where the pair model may be inadequate in any case.⁴⁰

III. EXPERIMENTAL DETAILS

We have determined the complex dielectric constant $\epsilon^* = \epsilon' - i4\pi\sigma/\omega$ from the shift in the resonator Q and resonant frequency (with respect to the case with no sample present) of a superconducting helical resonator⁴³ which spans the 0.1–2.1 GHz frequency range. The resonator was calibrated with a varactor diode used to span a set of known capacitance values and a niobium short circuit.^{44,45} Etched samples, 6-mm² area by 1-mm thick,

were inserted in the resonator capacitor with a gap maintained in series with the sample and one of the capacitor electrodes so as to prevent any spurious frequency dependence of $\sigma(\omega)$ arising from electrode effects.^{46,47} The gap capacitance was measured at 77 K where $4\pi\sigma/\omega\epsilon'$ $\gg 1.^{47,48}$ Any error in its determination results in a frequency-independent error in $\sigma(\omega)$ which does not alter the observed frequency dependence provided $4\pi\sigma/\omega\epsilon' \ll 1$. The power absorbed by the resonator was typically 10-500 pW and power increases by 10^3 or more resulted in negligible changes in $\epsilon^*(\omega)$. Si:P samples with donor densities in the range $(1.73-3.02) \times 10^{18}$ P atoms /cm³, determined from room-temperature resistivity measurements, were obtained from an ingot purchased from Pensilco Corp.; the upper limit on the density of compensating acceptors was $< 10^{14}$ cm⁻³ according to the manufacturer and the donor density was uniform to $\pm 2\%$. Another Si:P specimen with $N = 3.23 \times 10^{18}$ P atoms/cm³ was obtained from Cornell University.

Our rf source was an HP8753A network analyzer connected to an HP85046A S-parameter test set operating between 300 kHz and 3 GHz with bandwidths of 10 Hz to 3 kHz. This instrument, which incorporates a synthesized source and a sensitive detector, automatically measures the S-parameters (forward and reverse transmission and reflection coefficients) of the resonant cavity and has a "direct dump" feature which permits it to download all screen displays to a plotter via an IEEE-488 interface bus (giving us "real time" hard copy of all the resonance curves and parameters). rf power was coupled from the electronics measuring system to the resonator by inserting two coaxial lines through holes on the top of the resonator. Coupling through the top was used in order not to perturb the fields near the sample. Silverplated stainless-steel semirigid microcoaxial lines (Uniform Tubes UT85SS) were used to minimize thermal conduction from the measurement system to the resonator. The ends of the coaxial lines were formed into soldercoated probes. The probes provide coupling to the electric field and are advantageous as compared with magnetic coupling through loops because of their smaller size and the reduced cross talk between the two. They are ~ 1 cm long and the coupling to the resonator could be varied by changing their position within the coupling holes by means of an on-axis motor drive.

IV. RESULTS

A. Temperature dependence

For 2 < T < 4.2 K the conductivity was found to decrease as the temperature was lowered. Figure 1 shows a plot of the conductivity of the samples with $N = 1.73 \times 10^{18}$ and 2.47×10^{18} P atoms/cm³ as a function of 1/T for the 300, 900, 1100, and 2100 MHz harmonics (error bars on this and subsequent figures are approximately the size of the data points). For $N < 0.66N_c$, $\sigma(\omega, N, T)$ decreased with decreasing T by approximately a factor of 2 down to $T \sim 2$ K. For 1.5 K < T < 2 K, $\sigma(\omega, N, T)$ was independent of T within experimental error. The sample with $N = 2.47 \times 10^{18}$ P atoms/cm³ showed a much



FIG. 1. Temperature dependence of $\sigma(\omega, T)$ for $N = 1.73 \times 10^{18}$ (lower points) and $N = 2.47 \times 10^{18}$ P atoms/cm³ (upper points). Results are shown at four frequencies: solid triangles, 300 MHz; open triangles, 900 MHz; solid boxes, 1500 MHz; open boxes, 2100 MHz. Error bars are comparable to the size of the data points. The inset shows the helical resonator (cross hatched) with sample (solid), Teflon support for the capacitor (open), and coupling probes.

stronger temperature dependence and for our most concentrated samples, $N > 0.66N_c$, $\sigma(\omega, N, T)$ was weakly temperature dependent for 1.5 K < T < 4.2 K. The 100 and 300 MHz data were the most temperature dependent and, at the higher densities, still showed a residual (<15%) variation between 2 and 1.5 K. We now look at the frequency dependence of the temperature-dependent conductivity

$$\Delta \sigma(\omega, N, T) = \sigma(\omega, N, T) - \sigma_0 \omega^{s(N)}$$
$$= \sigma_1(N, T) \omega^{r(N, T)} ,$$

where $\sigma_0 \omega^{s(N)}$ is the nearly temperature-independent conductivity observed for T < 2 K to be discussed below. Figure 2 shows $\sigma_1(N,T)\omega^{r(N,T)}$ for three of our samples and shows that the residual frequency dependence of this term is nearly independent of temperature. This result, along with Fig. 1, justifies our separating the frequency and temperature dependences of $\sigma(\omega, N, T)$. We find r(N,T) > 1 for T > -2 K for the three lowest density samples: for $n = 1.73 \times 10^{18}$, $r = 1.19 \pm 0.07$; N = 1.91 $\times 10^{18}$, $r = 1.17 \pm 0.04$; $N = 2.10 \times 10^{18}$, $r = 1.05 \pm 0.10$. For the sample with $N = 2.47 \times 10^{18}$ P atoms/cm³ $r(N,T) \sim 0.7$. A roll off from the $\omega^{r(N,T)}$ behavior at the highest frequencies may be due to large values of the resonance prefactor I_0 : As discussed in the theoretical sec-



FIG. 2. Temperature-dependent conductivity $\sigma_1(N,T)\omega^{r(N,T)}$. Densities are in units of 10¹⁸ P atoms/cm³. Error bars are comparable to the size of the data points.

tion, phonon-assisted hopping processes involve^{17,20,40} two new hopping distances $r_{\omega} = (\xi/2) \ln(\nu_p/\omega)$ and $r_T = (\xi/2) \ln(2I_0/kT)$ where ν_p is a typical phonon frequency. If I_0 becomes large enough so that $\omega > \nu_p (kT/2I_0)^2$ then a much weaker frequency dependence is expected.^{20,22,40,48-50} Estimates of this term show that this could be our case at the lower densities: using $\nu_p = 10^{13}$ Hz, values of $2I_0/h$ calculated in the low-density limit¹⁴ and at T = 1.5 K, we estimate that the above inequality holds for $\nu > 1$ GHz for $N = 2.10 \times 10^{18}$ P atoms/cm³, the sample in which the roll off is most pronounced.

The exponents r(N, T) that we measure in Si:P are in good agreement with previous results at comparable reduced densities in Si:As.^{14,51} Figure 3 shows a plot of the average r(N, T) as measured in these systems between 4.2 and 2 K. r(N,T) decreases with increasing density towards values less than 1. The lower-density r(N,T) > 1values cannot be discussed in detail as the overall temperature dependence was small, as shown in Fig. 1, and thus the distinction between temperature-dependent σ_1 and temperature-independent σ_0 behavior may not be appropriate. The values of r(N,T) < 1 observed at the higher densities suggest a phonon-assisted hopping process. Α frequency-dependent conductivity with r(N, T) < 1 yields, through the Kramers-Kronig relations, a dielectric constant which diverges as ω^{r-1} at low frequencies. We do observe such an effect at the higher temperatures and this has been seen in Si:As (Ref. 14) as well.

The large temperature dependence of σ for the



FIG. 3. Exponent r(N,T) of the temperature-dependent conductivity in Si:P and Si:As. Solid triangles, this work; boxes, Ref. 14.

 $N = 2.47 \times 10^{18}$ P atoms/cm³ sample is consistent with experiments¹⁴ done on Si:As which showed a strong temperature dependence between 4.2 and 2 K at the higher dopant densities. We fit the temperature-dependent $\sigma_1(N,T)$ term for this sample to an Arrhenius-type law $\sigma_1(N,T) = \sigma'_1(N)e^{-(E_{act}/kT)}$. We could not fit this data to a power law of the type T^m unless large values of m are used. However, we could not rule out other mathematical fits such as a law of the type $Tme^{-(E/kT)}$. Si:As data in this temperature range was successfully fit^{14,51} with the use of an Arrhenius-type law. For this sample we found $E_{\rm act}$ to be independent of frequency and equal to 1.9±0.1 meV. We believe that we may associate $E_{\rm act}$ with the energy of the ε_2 mechanism which involves the excitation of the electrons from the Fermi level to the mobility edge; $\varepsilon_2 = E_c - E_F$. This process has been observed in Si:P (Refs. 52 and 53) and Ge:As (Refs. 54 and 55) near the metal-insulator transition. The activation energy that we measure at $N/N_c = 0.66$ is plotted in Fig. 4 along with other Si:P (Ref. 53) data and the results for Si:As.^{14,51} Our result fits very well with the previous data and we see that ε_2 decreases with increasing density, as expected, indicating an increase in the overlap between electronic states.5



FIG. 4. Activation energy $E_{\rm act}$ for Si:P and Si:As. Solid triangles, Si:P, Ref. 53 except for our point at $N/N_c = 0.66$; boxes, Si:As, Refs. 14 and 51. Error bars are comparable to the size of the data points.

B. Low-temperature conductivity: Frequency dependence

For temperatures between 1.5 and ~ 2 K we observed for all the samples a nearly temperature-independent conductivity $\sigma(\omega) = \sigma_0 \omega^{s(N)}$ as shown in the low-temperature region of Fig. 1. We stress, as was done before,¹⁶ that the Hess *et al.*²⁴ 400-MHz Si:P data showed a substantial temperature variation in the 20–120 mK range of the Mott form

$$\sigma(\omega, T) = \sigma_0(\omega) + \sigma_1 \exp(-(T_0/T)^{1/4})$$

which for small T_0 values would yield a relatively flat temperature dependence in the temperature range 1.4 K < T < 2 K. Thus the flat-plateau-like $\sigma(\omega)$ in this range should not necessarily be associated with the T=0K conductivity $\sigma(\omega, T=0)$. Similar behavior was observed in Si:As (Ref. 14) and we show in Fig. 5 a plot of $\sigma_0 \omega^{s(N)}$ versus frequency for three Si:P and three Si:As (Ref. 14) samples at comparable values of the reduced density N/N_c . The Si:P data shows the following behavior.

For frequencies between 100 MHz and 2.1 GHz and $N > 0.46N_c$, $\sigma(\omega)$ shows a frequency dependence with s(N) > 1. Moreover the $\sigma(\omega)$ versus ω curves get steeper as the donor density increases, indicating an *increase* of s(N) with N from s = 0.96 at $N = 1.73 \times 10^{18}$, to $s = 1.46\pm0.05$ at $N = 2.47 \times 10^{18}$, to $s = 2.20\pm0.09$ at $N = 3.23 \times 10^{18}$. The data between 0.1 and 2.1 GHz, when compared with the Si:P infrared data^{26,30} for $600 < v < 15\,000$ GHz, seem to be consistent in terms of



FIG. 5. Frequency dependence of the low-temperature conductivity $\sigma_0(N)\omega^{s(N)}$. Densities are in units of 10¹⁸ P atoms/ cm³. Si:P, this work; Si:As, Ref 14. Error bars are comparable to the size of the data points.

the frequency dependence for comparable values of N/N_c , as shown in Fig. 6, even though there is a gap with no data of a factor of 300 in frequency. We note the infrared data also yield s(N) > 2 for $N/N_c \sim 0.32$, 0.43, and 0.51 as v approaches the peak value of $\sigma(\omega, N)$ [σ_{\max} occurs at $v \approx 9000$ GHz], corresponding approximately to the position of the 1s $A_1 \rightarrow 2p$ donor transition in the dilute limit. Nevertheless the sharper frequency dependence observed as one approaches an absorption peak associated with intradonor transitions should not be directly compared with the frequency dependence observed at frequencies three orders of magnitude smaller.

The increase of s(N) with dopant density partially agrees with the Si:As results¹⁴ (Fig. 7) which show an increasing frequency exponent for $N/N_c < 0.66$, but disagrees with DC (Ref. 14) for $N/N_c > 0.66$. This discrepancy in s(N) for Si:P as compared to Si:As is not at present understood. Mott's³¹ theory [Eq. (1)] predicts s(N) decreasing with increasing dopant density due to the scaling behavior of the prefactor I_0 of the resonance enrgy⁵⁶ $I(r) = I_0 \exp(-r/\xi)$ included in his theory. Our data are apparently taken in a region where electronelectron interactions are the dominating energy: $e^2/\epsilon r_{\omega}$ $(\sim 30 \text{ K}) \gg kT (\sim 2 \text{ K}) \gg \hbar\omega$ (<0.1 K). Although DC (Ref. 14) argued that the frequency dependence with s(N) > 1 observed for $N/N_c < 0.75$ seemed to rule out the SE (Ref. 22) theory, we currently believe that our results permit us to test the SE (Ref. 22) prediction for the T=0zero-phonon hopping conductivity including these interactions Eq. (2). The reason for this is that, as shown in the discussion of Eq. (2) above, the SE (Ref. 22) prediction *qualitatively* predicts an exponent s(N) that increases with N. Simple estimates of the Coulomb energy term in Eq. (2) using the scaling behavior of ξ and I_0 and experimentally measured values of ϵ result in $e^2/\epsilon r_{\omega}$ being at least 30 times greater than $\hbar\omega$ (for the most concentrated sample at 2 GHz). Equation (2) for $\sigma(\omega)$ would then predict a frequency-dependent conductivity with s(N) < 1 for all our samples. Nevertheless, if the term $E_c = e^2 / \epsilon r_{\omega}$ is rewritten¹⁶ in terms of the characteristic



FIG. 6. Comparison of out data with infrared results. Densities are in units of 10^{18} P atoms/cm³. Optical data are from Refs. 26 and 30.



FIG. 7. Exponent s(N) of the low-temperature conductivity in Si:P and Si:As. Solid triangles, this work; boxes, Ref. 14. Note the partial overlap of the data at $N/N_c = 0.66$.

temperature T_0 of the Mott VRH one obtains

$$E_{c} = \frac{kT_{o}}{225} \left[\frac{\epsilon(N) - \epsilon_{h}}{\epsilon(N)} \right] \ln^{-1} \left[\frac{2I_{0}}{\hbar\omega} \right].$$
(3)

The Hess et al. experiments on Si:P (Ref. 24) at 400 MHz show that for a sample with $N = 3.1 \times 10^{18}$ P atoms/cm³, $T_0 = 15$ K. Interpolating their T_0 data on four different samples, for our sample with N = 3.02 $\times 10^{18}$ P atoms/cm³, we obtain $T_0 = 20$ K. Inserting T_0 into Eq. (3) we now have a Coulomb energy which is equal to the electromagnetic energy term (4 μ eV) at 1 GHz and smaller than this term at higher frequencies. It is possible to fit the low-temperature conductivity of this sample to the SE equation with $T_0 = 20$ K and $2I_0/h = 600$ GHz. We are also able to attempt to fit the samples with $N = 2.47 \times 10^{18}$ P atoms/cm³ ($T_0 = 50$ K, $2I_0/h = 1000$ GHz), and $N = 3.23 \times 10^{18}$ P atoms/cm³ $(T_0 = 13 \text{ K}, 2I_0 / h = 250 \text{ GHz})$, as shown in Fig. 8. The $N = 3.02 \times 10^{18}$ P atoms/cm³ sample shows a reasonable fit to Eq. (2). The fit for the highest-density sample has a smaller slope (1.85) than the data (2.20); this is because Eq. (2) inherently predicts s(N) < 2 and the fit shown is the best we could obtain. Attempts to fit the lowerdensity samples were unsuccessful because of the high values of the estimated T_0 's, resulting in a dominance at all frequencies of E_c over $\hbar\omega$ and thus giving a sublinear frequency dependent conductivity. We emphasize that the values of T_0 determined by Hess et al. at 400 MHz are very much smaller than T_0 values determined from dc-conductivity results for both Si:P (Refs. 3, 4, and 57) and Si:As (Ref. 1) for comparable values of N/N_c and for $(1 - N/N_c) > 0.1$. Thus, when we characterize $E_c \propto T_0$, as described earlier,¹⁶ and then employ the much smaller 400 MHZ values of T_0 we can semiquantitatively explain the frequency dependence of $\sigma(\omega, 1.5 < T < 2 \text{ K}) \propto \omega^{s(N)}$ and the density dependence of s(N), at least for $N/N_c > 0.66$. However, the origin of a strongly frequency-dependent T_0 is not understood and, as discussed earlier¹⁶ we are not aware of any VRH theory at finite frequencies that yields a frequency-dependent T_0 .



FIG. 8. Fits of the low-temperature conductivity to Eqs. (2) and (3) for three samples. Densities are in units of 10^{18} P atoms/cm³. Error bars are comparable to the size of the data points.

C. Low-temperature conductivity: Density dependence

The low-temperature conductivity $\sigma_0(N)\omega^{s(N)}$ discussed above showed the following density dependence: As a function of frequency, $\sigma(\omega, N)$ spans more than an order of magnitude at the lowest density and 2 orders of magnitude for the most concentrated sample, as the frequency changes by approximately a factor of 20. As a function of density the conductivity varies by a factor of 10^3 at 100 MHz and factor of 10^4 at 2 GHz, when the dopant density changes by less than a factor of 2. A strong density-dependent conductivity was observed in Si:As (Ref. 14) as well. Figure 9 shows a plot of the lowtemperature conductivity versus density for the 300, 1100, and 1900 MHz harmonics. Figure 10 compares our results with Si:As (Ref. 14) at 1 GHz and 1.5 K and shows that the systems behave in very similar manners. The density dependence in Si:As was generally found to be slightly stronger than in Si:P. Differences in the overall magnitude of the conductivity might be attributed to different compensation levels for the two systems. Equations (1) and (2) include density dependences.

(i) Through the density of states at the Fermi level, $N(E_F)$, which, in a free-electron model, is predicted to go as $N^{1/3}$. This is a weak power dependence with respect to our measurements. Specific-heat measurements on Si:P (Ref. 53) show that the density of states decreases as the carrier density decreases: The density of states at the Fermi level is given by $N(E_F)=(3/\pi^2k^2)\gamma$ where γ is the coefficient of the linear term of the temperature-dependent specific heat. From the c_v data $N(E_F)$ follows



FIG. 9. Density dependence of the conductivity. Densities are in units of 10^{18} P atoms/cm³. Error bars are comparable to the size of the data points.

free-electron behavior down to $N \approx 2.6 \times 10^{18}$ P atoms/cm³, below which it drops with decreasing density much faster than $N^{1/3}$, the free-electron behavior. Except for our two highest-density specimens, all of our samples fall in a density range where this data shows that $N(E_F)$ is much lower than the free-electron value and,



FIG. 10. Density dependence of the low-temperature conductivity at 1 GHz and 1.5 K. Solid triangles, Si:P, this work; boxes, Si:As, Ref. 14. Error bars are comparable to the size of the data points.

more importantly to the present discussion, a rapidly varying function of N. The rapid drop in $N(E_F)$ may represent the opening of a Coulomb gap suggested by the rapid increase in the Mott VRH characteristic temperature with decreasing density as observed in Si:P (Refs. 3, 4, and 57) and Si:As (Ref. 1) for $N < 0.9N_c$.

(ii) Through the localization length ξ which is expected to scale and diverge as N increases towards N_c . $\xi(N)$ is included in Eq. (2) as ξ^5 and, because of its scaling behavior, has a much stronger effect on the density dependence of the conductivity than the density of states discussed above.

(iii) Through the resonance integral prefactor I_0 , which is also expected to scale and decrease with increasing density. As discussed in detail by DC,¹⁴ the change in $I_0(N)$ is much too slow to explain the density dependences of s(N) and $\sigma(N)$ that we observe.

We have calculated the density dependence of the conductivity from the Shklovskii-Efros expression Eq. (2) using the electron-electron interaction term written¹⁶ as a function of T_0 . The resonance integral prefactor I_0 was calculated in the low density limit,¹⁴ the density of states at the Fermi level $N(E_F)$ was obtained from an interpolation of the specific-heat data⁵³ mentioned above and two values of the scaling exponent v, 1 and 0.5, were used. It is worth repeating that if $E_c \ll \hbar \omega$ then we expect a stronger N dependence at the higher frequencies because E_c , which is density dependent mainly through the factor ξ^5 , plays a lesser role in the calculation of σ . If, on the other hand, $\hbar\omega \ll E_c$, then the slope of the σ versus N curves will increase at higher N because electronelectron interactions will be smaller (at fixed frequency, due to the scaling behavior of T_0) than the electromagnetic energy. Our calculated conductivities exhibit these characteristics: The slope of $\sigma(N)$ versus N is higher at the higher densities. The main contribution to the increase of the slope at high densities comes from the localization length ξ , which is included in Eq. (2) as ξ^5 . The theoretical calculation qualitatively explains our data at the lower densities using a critical exponent v=1 but does not predict a density dependence as strong as we measure at the higher densities. If a critical exponent v=0.5 is used in the localization length, the density dependence predicted by Eq. (2) is much too slow to explain our data.

Equation (2) is valid only for frequencies much less than the crossover frequency ω_c . The apparent continuous increase of the predicted conductivity as $N \rightarrow N_c$ is not real, as ω_c is scaling to zero at the same time (see the theoretical discussion above) with the result that in our frequency range for densities very close to N_c Eq. (2) is not valid. The density dependence of $\sigma_0(N)\omega^{s(N)}$ is only partially explained by the SE theory through the factors $N(E_F)$, $\xi(N)$, and $I_0(N)$ in Eq. (2). However, the agreement is much better for $\nu = 1$ than for $\nu = \frac{1}{2}$. The lack of good agreement may represent the breakdown of the pair approximation in this regime as suggested by various authors.^{29,30} The importance of multielectron-correlated hopping^{58,59} may be essential in explaining the density dependence of $\sigma_0(N)\omega^{s(N)}$ in the critical-density regime.

V. SUMMARY AND CONCLUSIONS

In summary we have measured the conductivity of insulating Si:P with donor density spanning the range $0.46 < N/N_c < 0.86$, in the microwave range 0.1 < v < 2.1GHz and for temperatures between 4.2 and 1.5 K.

We have observed a nearly temperature-independent conductivity which shows a frequency dependence between 0.1 and 2.1 GHz of the type $\sigma(\omega) \propto \omega^{s(N)}$ with s(N) > 1.Over the entire density regime (0.46 $< N/N_c < 0.86$) of the present results the frequencydependence exponent s(N) increases with density. These results are not consistent with the T=0 K theoretical prediction of Mott nor are they consistent with the Shklovskii and Efros prediction using expected values of parameters entering the pair Coulomb energy E_c . If, however, the Coulomb energy E_c is substantially reduced, as suggested by the 400 MHz results of Hess et al. using the relation $E_c \propto T_0$ (with T_0 the Mott characteristic temperature), the behavior of s(N) can be explained for $N/N_c > 0.66$. The $\sigma_0 \omega^s$ low-temperature conductivity showed a very strong density dependence, the behavior of which depends on the harmonic frequency. Highdensity dependences of this type have been observed also in Si:As (Ref. 14) in this density and frequency range. the Shklovskii-Efros²² theory for the T = 0 K ac conductivity including electron-electron interactions cannot explain this strong density dependence over the entire density range of our measurements even if a critical exponent vof the localization length $(\xi = \xi_0 | 1 - N / N_c |^{-\nu})$ equal to 1 is used. v=1 is predicted by the scaling theories of localization but dc conductivity measurements on uncompensated doped semiconductors suggest v=0.5. Other properties, such as the divergence of the dielectric constant on the insulating side, are better explained by v=1. Our results furnish more information on the ac conduction processes in barely insulating doped semiconductors. As for Si:As,¹⁴ we measure strongly density-dependent conductivities which show superlinear frequency-dependent behavior $[\sigma(\omega, N) \propto \omega^{s(N)}, s(N) > 1]$ at low temperatures. The main novelty of the present work is the apparent continuous increase of the conductivity exponent s(N)with dopant density. The Si:As conductivity exponent s(N) showed¹⁴ a turnaround in its behavior at $N/N_c > 0.66$. At the present time we are unable to reconcile these different results. A new set of experiments, in both systems, should be undertaken and extended to much lower temperatures so that measurements closer to N_c can be made.

The helical resonator is a unique tool to further these studies because it spans a wide frequency range and thus permits ac conductivities to be measured without multiple temperature cyclings needed with conventional resonant cavity measurements. The advantage of obtaining lower-temperature measurements has been made clear by the present work. Dilution-refrigerator experiments may answer the following questions.

(i) Is the nearly temperature-independent behavior of $\sigma(\omega)$ observed for $T < \sim 2$ K a plateau connecting the Mott variable-range hopping law observed by Hess *et al.*²⁴ at 400 MHz and 20 < T < 120 mK and the activated behavior which we observe for T > 2 K? (This point raises the added question of the possibility of extending these measurements to *higher* temperatures up to the critical temperature for Niobium. In so doing we may better document the activated behavior of $\sigma(\omega)$ for T > 2 K and try to discriminate between the possible temperatures dependences of which the Arrhenius law is only one example.)

(ii) Having made¹⁶ the connection between the magnitude of electron-electron interactions E_c and the Mott variable-range-hopping characteristic temperature T_0 , we used this result, along with the measured²⁴ values of T_0 at 400 MHz, to semiquantitatively explain the apparent continuous increase of s(N) with dopant density. In doing this we have shown an important aspect of the VRH process: T_0 appears to be frequency dependent and a decreasing function of frequency. The helical resonator is again the optimal tool to further study this. By spanning a factor of 20 or more in frequency and doing measurements in the mK range, this device could help document any frequency dependence of T_0 .

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mun. 30, 661 (1979).

- ⁴Y. Ootuka, F. Komori, Y. Monden, S. Kobayashi, and W. Sasaki, Solid State Commun. **33**, 793 (1980).
- ⁵D. W. Koon and T. G. Castner, Solid State Commun. **64**, 11 (1987).
- ⁶M. Paalanen, T. F. Rosenbaum, G. A. Thomas, and R. N. Bhatt, Phys. Rev. Lett. 48, 1284 (1982).
- ⁷P. F. Newman and D. F. Holcomb, Phys. Rev. B 28, 628 (1983).

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¹W. N. Shafarman and T. G. Castner, Phys. Rev. B 33, 3570 (1986).

²W. N. Shafarman and T. G. Castner, in Proceedings of the Seventeenth International Conference on the Physics of Semiconductors, San Francisco, 1984, edited by D. J. Chadi and W. A. Harrison (Springer-Verlag, New York, 1985), p. 1079.

³S. Kobayashi, Y. Monden, and W. Sasaki, Solid State Com-

- ⁸N. F. Mott, J. Non-Cryst. Solids 1, 1 (1968).
- ⁹A. L. Efros and B. I. Shklovskii, J. Phys. C 8, L49 (1975).
- ¹⁰M. Benzaquen and D. Walsh, Phys. Rev. B 30, 7287 (1984).
- ¹¹F. R. Allen and C. J. Adkins, Philos. Mag. 26, 1027 (1982).
- ¹²D. Redfield, Phys. Rev. Lett. **30**, 1319 (1973).
- ¹³A. N. Ionov, I. S. Shlimak, and M. N. Matveev, Solid State Commun. 47, 763 (1983).
- ¹⁴R. J. Deri and T. G. Castner, Phys. Rev. Lett. 57, 134 (1986).
- ¹⁵T. G. Castner and R. J. Deri, in *Disordered Semiconductors*, edited by M. A. Kastner, G. A. Thomas, and S. R. Ovshinsky (Plenum, New York, 1987), p. 73.
- ¹⁶M. Migliuolo and T. G. Castner, Solid State Commun. 67, 863 (1988).
- ¹⁷M. Pollak and T. H. Geballe, Phys. Rev. **122**, 1742 (1961).
- ¹⁸S. Golin, Phys. Rev. 132, 178 (1963).
- ¹⁹H. S. Tan and T. G. Castner, Phys. Rev. B 23, 3983 (1981).
- ²⁰I. G. Austin and N. F. Mott, Adv. Phys. 18, 41 (1969).
- ²¹M. Paalanen, T. F. Rosenbaum, G. A. Thomas, and R. N. Bhatt, Phys. Rev. Lett. **51**, 1896 (1983).
- ²²B. I. Shklovskii and A. L. Efros, Zh. Eksp. Teor. Fiz. 81, 405 (1981) [Sov. Phys.—JETP 54, 218 (1981)].
- ²³T. G. Castner, W. N. Shafarman, R. J. Deri, and J. S. Brooks,
 J. Phys. C 19, 491 (1986).
- ²⁴H. F. Hess, K. DeConde, T. F. Rosenbaum, and G. A. Thomas, Phys. Rev. B 25, 5578 (1982).
- ²⁵S. Tanaka and Y. Fan, Phys. Rev. 132, 1516 (1963).
- ²⁶R. C. Milward and L. J. Neuringer, Phys. Rev. Lett. **15**, 664 (1965).
- ²⁷S. Toyotomi, J. Phys. Soc. Jpn. 38, 175 (1975).
- ²⁸P. Townsend, J. Phys. C 11, 1481 (1978).
- ²⁹M. Capizzi, G. A. Thomas, F. De Rosa, R. N. Bhatt, and T. M. Rice, Phys. Rev. Lett. 44, 1019 (1980).
- ³⁰G. A. Thomas, M. Capizzi, F. DeRosa, R. N. Bhatt, and T. M. Rice, Phys. Rev. B 23, 5472 (1981).
- ³¹N. F. Mott, Philos. Mag. 22, 7 (1970).
- ³²B. Shapiro and E. Abrahams, Phys. Rev. B 24, 4889 (1981).
- ³³Y. Imry, Y. Gefen, and D. J. Bergam, in Anderson Localization, Proceedings of the 4th Taniguchi International Symposium, Sanda-Shi 1981 Solid State Science Series (Vol. 39)

(Springer-Verlag, Berlin, 1982), p. 138.

- ³⁴P. Wolfe and D. Vollhart, in Anderson Localization, Proceedings of the 4th Taniguchi International Symposium, Sanda-Shi 1981 Solid State Science Series (Vol. 39) (Springer-Verlag, Berlin, 1982), p. 26.
- ³⁵R. N. Bhatt, Philos. Mag. **B50**, 189 (1984).
- ³⁶W. Gotze, J. Phys. C 12, 1279 (1979).
- ³⁷E. Abrahams and P. A. Lee, Phys. Rev. B 33, 683 (1986).
- ³⁸N. F. Mott and M. Kaveh, Philos. Mag. B52, 177 (1985).
- ³⁹N. F. Mott and E. A. Davis, *Electronic Processes in Non Crystalline Materials* (Clarendon Press, Oxford, 1979).
- ⁴⁰H. Bottger and V. V. Bryskin, Phys. Status Solidi 78, 415 (1976).
- ⁴¹R. N. Bhatt and T. V. Ramakrishnan, J. Phys. C 17, L639 (1984).
- ⁴²R. N. Bhatt, Philos. Mag. B50, 189 (1984).
- ⁴³R. J. Deri, Rev. Sci. Instrum. 57, 82 (1986).
- ⁴⁴M. Migliuolo and R. J. Deri, Rev. Sci. Instrum. 58, 892 (1987).
- ⁴⁵R. J. Deri and M. Migliuolo, Rev. Sci. Instrum. 58, 890 (1987).
- ⁴⁶X. LeCleac'h, J. Phys. (Paris) 40, 417 (1979).
- ⁴⁷R. A. Street, G. R. Davies, and A. D. Yoffe, J. Non-Cryst. Solids 5, 276 (1971).
- ⁴⁸S. Tanaka, M. Kobayashi, E. Hanamura, and K. Uchinokura, Phys. Rev. **134**, A256 (1964).
- ⁴⁹M. Pollak, Phys. Rev. 133, A564 (1964).
- ⁵⁰M. Pollak, Philos. Mag. 23, 519 (1971).
- ⁵¹R. J. Deri, Ph.D. thesis, University of Rochester, 1986.
- ⁵²C. Yamanouchi, K. Mizuguchi, and W. Sasaki, J. Phys. Soc. Jpn. 22, 859 (1967).
- ⁵³N. Kobayashi, S. Ikehuta, S. Kobayashi, and W. Sasaki, Solid State Commun. 24, 67 (1977).
- ⁵⁴H. Fritzche, Phys. Rev. 119, 1899 (1960).
- ⁵⁵E. A. Davis and W. D. Compton, Phys. Rev. **140**, A2183 (1965).
- ⁵⁶A. Miller and E. Abrahams, Phys. Rev. 120, 745 (1960).
- ⁵⁷W. Sasaki, Philos. Mag. **B52**, 427 (1985).
- ⁵⁸M. Pollak, Proc. R. Soc. London A325, 383 (1971).
- ⁵⁹M. Pollak and M. L. Knotek, J. Non-Cryst. Solids **32**, 141 (1979).