Hopping Conduction and Metal-Insulator Transition in Isotopically Enriched Neutron-Transmutation-Doped ^{70}Ge:Ga

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We report on the electrical conductivity σ of a series of nominally uncompensated neutron-transmutation-doped isotopically enriched ^{70}Ge:Ga samples with the Ga concentration [Ga] near N_c for the metal-insulator transition. σ of all insulating samples obeys $\ln\sigma \propto -(T_0/T)^{1/2}$ with $T_0 \propto (N_c\text{-[Ga]})/N_c$ while the zero temperature conductivity $\sigma(0)$ of the metallic samples is $\sigma(0) \propto \{([\text{Ga}]\text{-}N_c)/N_c\}^{\nu}$ with the critical exponent $\nu \approx 0.5$. The values of N_c obtained from the two independent scalings of T_0 and $\sigma(0)$ are identical, i.e., $\nu \approx 0.5$ is established unambiguously for uncompensated Ge:Ga. [S0031-9007(96)01533-5]

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In experimental studies of the metal-insulator (MI) transition one measures the critical behavior of physical quantities such as conductivity, dielectric constant, heat capacity, etc. The doping induced MI transition in semiconductors is considered to be a model case for the general theory of the critical behaviors of solids. In particular the conductivity extrapolated to zero temperature $[\sigma(0)]$ is evaluated routinely as a function of doping concentration (N) immediately above the MI transition critical concentration (N_c);

$$\sigma(0) = \sigma_0[(N - N_c)/N_c]^{\nu} \quad (1)$$

where σ_0 is the prefactor and ν is the critical exponent. The value of ν, determined experimentally, is compared with theoretical predictions. Up to now $\nu \approx 0.5$ has been obtained with nominally uncompensated semiconductors (Si:P [1], Si:As [2,3], Ge:As [4], Si:B [5]) while $\nu \approx 1$ has been found with compensated semiconductors (Ge:Sb [6], Si:P,B [7], Ge:Ga,As [8]) and amorphous alloys [9–12]. Exceptions are uncompensated Ge:Sb with $\nu \approx 1$ [13] and $\text{Ga}_x\text{Ar}_{1-x}$ amorphous alloys with $\nu \approx 0.5$ [14]. As we explain below, the value of $\nu \approx 0.5$ obtained with simple systems like uncompensated semiconductors turns out to be inconsistent with theoretical predictions [15]. In his original theory Mott considered only the electron-electron (e^--e^-) interaction (Mott transition) and predicted a discontinuous transition of $\sigma(0)$ at N_c [16]. Although there is much evidence for the importance of e^--e^- interactions, no experimental observation of such an abrupt transition has been reported. Anderson's idea of MI transitions is based solely on the disordered potential arising from randomly distributed dopants (Anderson transition) [17]. This lead to the development of the well-known "scaling theory" which predicted $\nu \approx 1$ for three dimensional systems [18]. More recently, higher order calculations of the scaling theory (exclusively with disorder and no interactions) predict $\nu \approx 1.3$ [19], and, more importantly, this value is shown to be independent of time reversal invariance [20] and of the strength of spin-orbit interactions [21]. It is therefore clear that the effect of disorder alone cannot explain the experimental results of $\nu \approx 0.5$ or 1. Chayes *et al.* combined the theories of Mott and Anderson and successfully set the lowest limit $\nu > 2/3$ [22]. This result permits $\nu \approx 1$ obtained with compensated semiconductors and amorphous alloys. However, there still is no theory which can convincingly explain $\nu \approx 0.5$ found for uncompensated semiconductors.

Very recently a different interpretation of results obtained with uncompensated Si:P leading to $\nu \approx 1.3$ has been proposed by Löhneysen and co-workers [23]. They fitted data for $\sigma(0) < 5\ (\Omega\ \text{cm})^{-1}$ samples with Eq. (1) by redefining N_c which was 6% smaller than the conventionally accepted value. Their results, which agree perfectly with the model of the disorder induced transition, led to many discussions especially among experimentalists. Rosenbaum, Thomas, and Paalanen argued that the $\nu \approx 1.3$ region below the conventional N_c should be ignored since it was most likely an artifact due to inhomogeneous dopant distribution [24]. Castner, on the other hand, suggested that the $\sigma(0) < 5\ (\Omega\ \text{cm})^{-1}$ samples in Ref. [23] are insulators since their resistivity ρ obeys Mott's variable range hopping theory characterized by $\ln\rho \propto T^{-1/4}$ [25].

The questions raised can be answered only if a series of homogeneously doped samples can be prepared which allows an unambiguous determination of N_c i.e., an appropriate scaling of the conductivity to N_c becomes possible from both the insulating ($N < N_c$) and the metallic ($N_c < N$) sides of the transition. In this Letter we

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describe such an experiment using nominally uncompensated *p*-type Ge:Ga semiconductors.

The present study is the result of an extensive effort to produce homogeneously doped samples near N_c. Even with today's advanced semiconductor technology, melt-doping of bulk semiconductors always leads to inhomogeneous dopant distributions due to impurity segregation and striation during crystal growth. We have overcome such problems by applying the neutron transmutation doping (NTD) technique to a chemically pure, isotopically enriched ^{70}Ge crystal. A similar approach using a limited number of NTD ^{74}Ge:As samples has been employed by Ionov *et al.* [4]. The ^{70}Ge crystal of isotopic composition $[^{70}\mathrm{Ge}] = 96.2$ at.% and $[^{72}\mathrm{Ge}] = 3.8$ at.% was grown using the Czochralski method developed for ultra-pure Ge [26]. The as-grown crystal was free of dislocations, *p*-type with an electrically active net-impurity concentration less than 5×10^{11} cm^{-3}. A number of wafers were cut from the ingot and irradiated with thermal neutrons at the University of Missouri Research Reactor. Upon capturing a thermal neutron ^{70}Ge becomes ^{71}Ge which decays via electron capture to a ^{71}Ga acceptor with a half-life of $T_{1/2} = 11.2$ days. A very small fraction of ^{72}Ge becomes ^{73}Ge which is stable, i.e., no other acceptors or donors are introduced. We use NTD since it is known to produce the most homogeneous, perfectly random dopant distribution down to the atomic level [27]. We have shown previously that the concentration $[^{71}\mathrm{Ga}]$ in our ^{70}Ge samples after NTD is given precisely by

$$[^{71}\mathrm{Ga}](\mathrm{cm}^{-3}) = 0.1155 \times n(\mathrm{cm}^{-2}), \qquad (2)$$

where n is the thermal neutron fluence [28]. An accurate control of the neutron fluence n permits us to achieve the desired concentrations $[^{71}\mathrm{Ga}]$. The number ratio of thermal to fast neutrons during NTD was $\sim 30:1$. It has been shown that the compensation ratio in our NTD ^{70}Ge:Ga becomes less than 0.001 after annealing at 650 °C for 10 sec [28]. The short annealing time is selected in order to avoid redistribution of the homogeneously distributed ^{71}Ga. The dimension of most samples for the conductivity measurements was $6 \times 2 \times 0.2$ mm^3. Four strips of boron ion implanted regions on a 6×2 mm^2 face of each sample were coated with 200 Å Pd and 4000 Å Au pads. Annealing at 300 °C for one hour activated the implanted boron and removed the stress in the metal films. 25 μm Au wires were bonded ultrasonically to the Au pads for four-point electrical measurements as well as for heat sinking. The two point measurements with the implanted contacts were employed for some of the high resistivity insulating samples. All low frequency (<30 Hz) conductivity measurements were performed in a dilution refrigerator using a standard lock-in amplifier and/or an ac bridge (RV-Elektroniikka, AVS-46). Sample heating was avoided by using an electrical power of less than 10^{-14} W.

Figure 1 shows the temperature (T) dependence of the resistivities (ρ) of 14 insulating samples in the range $N = 0.16$–$0.99N_c$. All curves become linear only when $\ln\rho$ is plotted against $T^{-1/2}$ in good agreement with Efros and Shklovskii's theory of variable range hopping conduction for strongly interacting electrons [29]:

$$\rho = \rho_0 \exp(T_0/T)^{1/2}, \qquad (3)$$

where ρ_0 is a prefactor and T_0 is given by

$$T_0 \approx 2.8e^2/\kappa(N)\xi(N). \qquad (4)$$

Here $\kappa(N)$ and $\xi(N)$ are the dielectric constant and localization length depending on N, respectively. Moreover, $\kappa(N) \propto [N_c/(N_c - N)]^s$ and $\xi(N) \propto [N_c/(N_c - N)]^\zeta$ as N approaches N_c from the insulating side so that T_0 becomes [29]

$$T_0 = A[(N_c - N)/N_c]^\alpha, \qquad (5)$$

where $\alpha = s + \zeta$ is to be determined experimentally. The left half of Fig. 2 shows the experimentally determined T_0 vs [Ga] (filled diamonds) together with the result of a three-parameter-fitting using A, N_c, and α as variables in Eq. (5) (solid curve). We deduced [Ga] for each sample using Eq. (2) since we know the precise neutron fluence used in each irradiation run. The best fit of T_0 with Eq. (5) was obtained with the values $\alpha = 1.03 \pm 0.038$ and $N_c = 1.855 \pm 0.012 \times 10^{17}$ cm^{-3}. A much larger value of $\alpha \approx 2$ has been reported for Ge:As using only three samples with the highest N being far from the transition ($0.56N_c$) [30]. T_0' of the Mott's variable range hopping theory [$\ln\rho \propto (T_0'/T)^{1/4}$] has also been scaled to N_c for Si:P and Si:As leading to a severe disagreement, $\alpha \approx 1.45$ for Si:P [31] and $\alpha \approx 2.9$ for Si:As

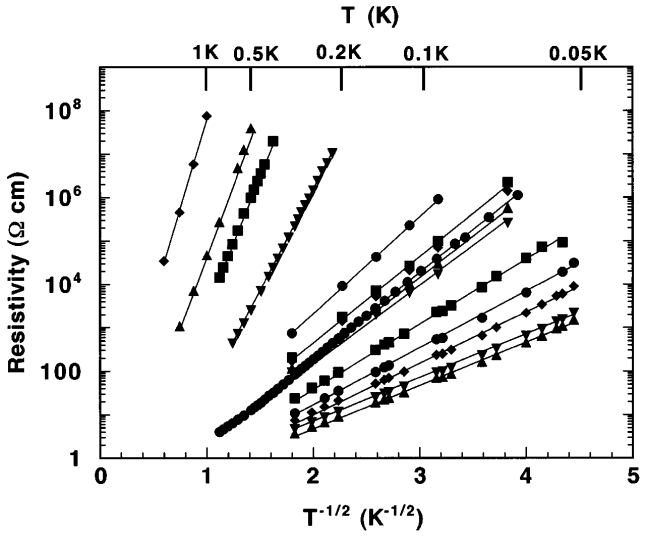

FIG. 1. The logarithm of the resistivity plotted as a function of $T^{-1/2}$ for 14 insulating NTD ^{70}Ge:Ga samples. Gallium concentrations from top to bottom in units of 10^{16} cm^{-3} are 3.02, 8.00, 9.36, 14.50, 17.17, 17.52, 17.61, 17.68, 17.70, 17.79, 17.96, 18.05, 18.23, and 18.40.

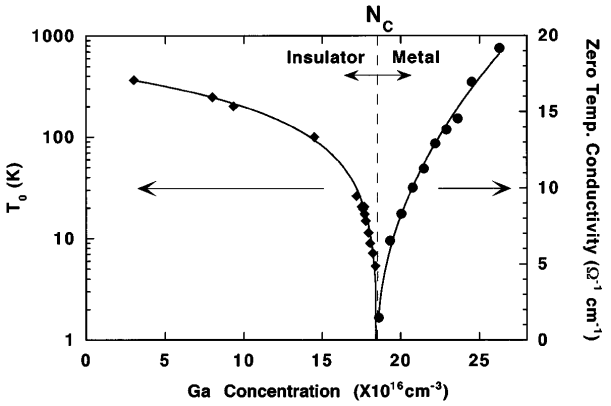

FIG. 2. The left side shows the experimentally determined T_0 of 14 insulating samples as a function of Ga concentration (◆). The solid curve is the best fit obtained with Eq. (5) with $\alpha \approx 1$. The right side shows the zero temperature conductivity $\sigma(0)$ obtained from the extrapolations in Fig. 3 for ten metallic samples as a function of Ga concentration (•). The solid curve is the best fit obtained with Eq. (1) with $\nu \approx 0.5$.

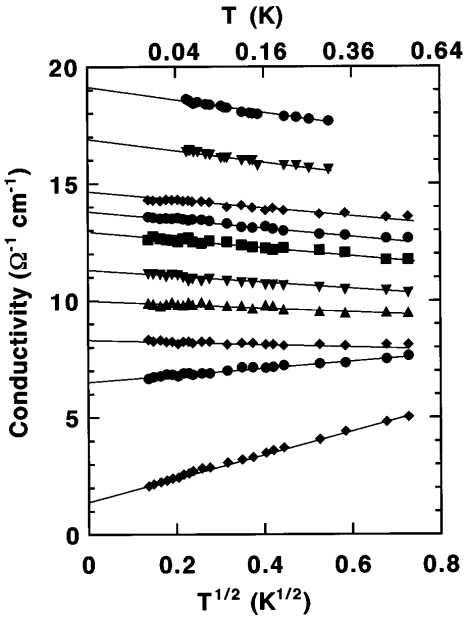

FIG. 3. Conductivity plotted as a function of $T^{1/2}$ for 10 metallic NTD ^{70}Ge:Ga samples. Solid lines indicate extrapolation to $T = 0$ K. Gallium concentrations from top to bottom in units of 10^{16} cm^{-3} are 18.61, 19.33, 20.04, 20.76, 21.47, 22.19, 22.90, 23.62, 24.50, and 26.25.

[32], even though these two systems are considered to be very similar. Inhomogeneous dopant distributions may have affected the results of these Si studies. In the present study, $\alpha = 1$ has been obtained with 14 homogeneously doped samples of [Ga] = 0.16-0.99N_c, all demonstrating the $\ln\rho \propto T^{-1/2}$ dependence, i.e., this data set should be considered to be the first reliable determination of the hopping conductivity exponent α for a particular semiconductor system. s for $\kappa(N)$ is another important exponent to be determined in the future for our NTD Ge. Hess *et al.* have found $s = 1.15$ and determined N_c accurately for Si:P [33].

Figure 3 shows the conductivity σ in ten metallic samples plotted against $T^{1/2}$. Our extrapolation of each curve to $T = 0$, i.e., the determination of the zero temperature conductivity $\sigma(0)$, yields a very small error since the dependence of σ on T for all samples is very weak. σ plotted against other functions of temperatures such as $T^{1/x}$ with $x = 1$–3 does not change significantly the values of $\sigma(0)$. The right half of Fig. 2 shows $\sigma(0)$ as a function of [Ga] (filled circles) together with a fit obtained by the scaling expression Eq. (1) (solid curve). The values of the parameters determined from this fit are $\nu = 0.502 \pm 0.025$ and $N_c = 1.856 \pm 0.003 \times 10^{17}$ cm^{-3}. Here we present $\nu \approx 0.5$ for uncompensated Ge:Ga semiconductors with high confidence, since the two values of N_c obtained from the scaling of T_0 [Eq. (5)] and $\sigma(0)$ [Eq. (1)] agree perfectly. The Karlsruhe group suggested that only the samples with $d\sigma/d\sqrt{T} > 0$ in Fig. 3 should be included in the fitting with Eq. (1) [23]. We have only two samples exhibiting such a behavior. However, with N_c fixed hard at 1.855×10^{17} cm^{-3} by the fitting with Eq. (5), $\nu \approx 0.5$ is obtained even with the $\sigma(0)$ of our two $d\sigma/d\sqrt{T} > 0$ samples. Therefore the present work lends strong support to $\nu \approx 0.5$ obtained previously with many nominally uncompensated semiconductors [1–5]. We agree with Rosenbaum, Thomas, and Paalanen that the $\nu \approx 1.3$ region observed by the Karlsruhe group is most likely an artifact due to an inhomogeneous dopant distribution [24], because we did not observe such a region with our homogeneously doped samples.

So far we have considered only the scaling of the zero temperature conductivity $\sigma(0)$ as a function of N. In the future it will be of great interest to evaluate the dynamical scaling behavior of the conductivity as a function of both T and N [Eq. (5.44b) of Ref. [15] by Belitz and Kirkpatrick]. We are in the process of fabricating new NTD Ge samples closer to N_c for such an analysis. Also it is important to note that the combination of Wegner's scaling law $\nu = \zeta$, the relation $s = 2\zeta$, and the experimentally determined $\nu \approx 0.5$ predicts $\alpha = s + \zeta \sim 1.5$ for our Ge:Ga. This value differs slightly from the experimentally determined $\alpha \approx 1$. Further investigations are underway to clarify this point.

In conclusion we have determined the critical exponent for uncompensated Ge:Ga to be $\nu \approx 0.5$. The homogeneous dopant distribution, we believe, has been the key for our successful experiment demonstrating the scaling behavior of the conductivity on both sides of the transition.

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