# Neutral impurity scattering in mercury-doped germanium

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A hole mobility analysis is described for weakly compensated mercury-doped germanium over a temperature range (10 < T < 100 K) for which neutral Hg acceptors were predominant and made a detectable contribution to hole scattering. It was deduced that  $\mu_N N_N = 4.4 \times 10^{20} \text{ cm}^{-1} \text{V}^{-1} \text{s}^{-1}$  from 10 to  $\sim 35 \text{ K}$ ; a scattering efficiency 40% of that to be expected for a shallow monovalent acceptor. The scattering efficiency appeared to halve between 35 and 50 K, and to continue at a rate consistent with  $\mu_N N_N = 8.8 \times 10^{20} \text{ cm}^{-1} \text{V}^{-1} \text{s}^{-1}$  from 50 K upwards. Thus even in the upper part of the temperature range, Hg<sup>0</sup> acceptors were twice as efficient in scattering as might have been supposed from the mercury center's first ionization energy. It is noted that the ratio of hole wavelength to interacceptor spacing was propitious for a contribution to scattering by acceptor pairs. That complication would not be easy to avoid experimentally.

# I. INTRODUCTION

Carrier mobilities in a moderately doped semiconductor are usually controlled by lattice-phonon scattering for high temperatures and by ionized impurity scattering for lower temperatures. Neutral impurity scattering is presumed to make some contribution, possibly detectable at low temperatures. However, since a neutral impurity may have only a few percent of the scattering efficiency of an ionized one, conditions under which neutral center scattering can dominate are not common.

This paper describes an analysis of hole transport for weakly compensated mercury-doped germanium, including allowance for neutral acceptor  $(Hg^0)$  scattering. Substitutional Hg is a divalent acceptor in germanium,<sup>1</sup> with successive ionization energies  $E_{a1}=91$  meV and  $E_{a2}=230$  meV. For thermal equilibrium below 200 K, the Fermi level  $E_F$  is low enough in weakly compensated material so that the Hg<sup>2-</sup> state will not occur. Moreover, Hg<sup>0</sup> are much more numerous than Hg<sup>-</sup> below 100 K. Thus Ge:Hg seemed potentially promising as a system in which the contribution of Hg<sup>0</sup> to hole scattering might be measurable over an extended temperature range.

The scattering of holes by  $Hg^0$  may be regarded as crudely analogous to the atomic physics situation of electron scattering by helium atoms.<sup>2,3</sup> The scattering of electrons and holes by neutral monovalent impurities has been modeled<sup>4-8</sup> on concepts of electron-hydrogen and positron-hydrogen scattering.

In the present work, conductivity and Hall-effect data—supplemented eventually with photoconductivity and photo-Hall data—were analyzed for the scattering contributions of phonons, ionized impurities, and neutral acceptors. These were evaluated separately for the heavy-hole and light-hole bands over the range 10-150 K.

The analysis indicated that for  $T \sim 20$  K, Hg<sup>o</sup> centers contributed up to 35% of the heavy-hole scattering and 75% of the light-hole scattering for material with  $N_a \simeq 10^{15}$  cm<sup>-3</sup> and <0.3% compensation. The scattering by Hg<sup>o</sup> played a lesser role for higher temperatures, partly because of increased lattice scattering, but also because of first-stage acceptor ionization. The ionization Hg<sup>o</sup>  $\rightarrow$ Hg<sup>-</sup> became substantial above 100 K, and Hg<sup>o</sup> scattering was not detectable by 150 K.

The search for Ge:Hg with appropriate amounts of mercury doping and compensation was enlivened by extraneous phenomena seen in several samples. These were consequences of spatial inhomogeneity of the effective compensator concentration. Those complications, which are discussed elsewhere,<sup>9</sup> dictated the choices of samples and of measurement techniques for mobility analyses.

### **II. MODELS FOR NEUTRAL IMPURITY SCATTERING**

A realistic model for hole scattering by a neutral divalent acceptor (such as Hg in Ge) should take into account interactions of the incident mobile carrier with the two bound holes. That will not be embarked upon here. The two bound holes for neutral Hg in Ge have an interesting Coulomb interaction that results in an acceptor ground state split<sup>10</sup> by some 0.7 meV.

Attempts to model scattering by neutral impurities have concentrated on the simpler situation of monovalent "effective mass" impurities, of ionization energy  $E_{\rm em} = e^4 m^* / 2\hbar^2 \kappa^2$ , and ground-state wave function radius  $a_{\rm em} = \kappa \hbar^2 / 2e^2 m^* = e^2 / 2\kappa E_{\rm em}$  $= (\hbar^2 / 2m^* E_{\rm em})^{1/2}$ . While such models are not directly applicable to the Ge:Hg situation, a brief summary is useful here, inasmuch as experimental data are invariably traced back to a comparison

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with what a "hydrogenic" impurity is expected to do.

The first such model was that of Erginsoy.<sup>4</sup> He used the analogy with slow electron scattering by atomic hydrogen<sup>11</sup> and concluded that only spherically symmetric scattering (zero-order phase shift  $\delta_0$ ) need be considered for an incident electron of kinetic energy less than  $0.25E_{\rm em}$ . That led to a speed-independent relaxation time  $\tau = (m^* e)^2/20\kappa\hbar^3N_N$ , and to a temperature-independent "Erginsoy mobility"

$$\mu_{E} = m^{*} e^{3} / 20 \kappa \hbar^{3} N_{N} = e / 20 \hbar a_{\rm em} N_{N} = E_{\rm em} \kappa / 10 e \hbar N_{N} .$$
(1)

Equation (1) is expressed in three forms, since a mobility  $\mu_N$  which differs from  $\mu_E$  can be parametrized by substitution of an appropriately "scaled" effective mass,<sup>12</sup> or by adjustment of the effective radius  $a_{\rm em} + a^*$ , or of the ionization energy  $E_{\rm em} + E^*$ . Equivalently, one may simply write  $\mu_N = A\mu_E$ , with the scaling described by a numerical ratio.<sup>13</sup>

Models since Erginsoy's have continued to concentrate on hydrogenic analogies. Ansel'm<sup>5</sup> noted that atomic hydrogen forms a weakly bound H<sup>-</sup> system and reasoned thereby that scattering by neutral hydrogenic impurities has a resonant component. This led to a speed-dependent  $\tau$  and thus to a temperature-dependent  $\mu_N(T)$ . Subsequent models of Sclar,<sup>6</sup> Blagosklonskaya *et al.*,<sup>7</sup> and McGill and Baron<sup>8</sup> predict varying degrees of temperature dependence for  $\mu_N$ , though all with magnitude generally similar to  $\mu_E$  of Eq. (1). However, McGill and Baron showed that  $\tau$  should rise substantially for carrier kinetic energy less than 1% of the impurity ionization energy, which should boost  $\mu_N$ significantly for very low temperatures.

McGill and Baron<sup>8</sup> also commented on the scaling to be expected for  $\mu_N$  when the neutral centers are non hydrogenic and of ionization energy  $E_B > E_{em}$ . One may then expect<sup>14</sup> a bound state wave-function of radius  $a_B = a_{em} (E_{em}/E_B)^{1/2}$ , whether the effective potential is essentially Coulomb  $[\psi(r) \sim \exp(-r/a_B)]$ or more nearly short range in character  $[\psi(r) \sim r^{-1}\exp(-r/a_B)]$ . Assuming that the scattering cross section is proportional to  $a_B^2$ , then the mobility  $\mu_N = E_B \kappa / 10e\hbar N_N = \mu_E E_B / E_{em}$ . Baron *et al.*<sup>15</sup> show that neutral scattering data for silicon doped with As, Ga, or In has this trend of  $\mu_N$  with  $E_B$ .

Thus one convenient way to incorporate crosssection scaling for a deep or multivalent impurity, and also the temperature dependence to be typically expected for  $\mu_N$ , is through use of a quantity  $E^*(T)$ with dimensions of energy. If that quantity is expressed in electron volts, then

$$\mu_{N} = \mu_{E} E^{*} / E_{em} = E^{*} \kappa / 10 e \hbar N_{N}$$
  
= 1.055 × 10<sup>21</sup> E<sup>\*</sup> \kappa / N\_{N} cm<sup>2</sup> / V s. (2)

The results of the present work with Ge:Hg are quoted in Sec. V in terms of  $\mu_N N_N$  and of  $E^*$  for neutral mercury. Since Hg is divalent rather than monovalent, a close correspondence between  $E^*$  and  $E_{a1}$  should not be presumed. The data actually indicate an increase of  $E^*$  (decrease of scattering efficiency) with rising temperature, but with  $E^* < E_{a1}$  for all temperatures observable.

#### **III. SOME PREVIOUS EXPERIMENTAL RESULTS**

Much of the published experimental literature has been for monovalent impurities. Norton et al.<sup>16</sup> analyzed mobility data for silicon containing Group V donors, and extracted data of  $\mu_N$  over the range 6-100 K. For each kind of donor,  $\mu_N N_N$  showed a shallow minimum around 6 K and a shallow maximum around 50 K. For  $T \simeq 30$  K, the neutral scattering rates were consistent with  $E^* \simeq 50$  meV for either P or Sb donors (comparable with  $E_d$  for those donors). However, As donors were much less efficient in scattering ( $E^* \simeq 170$  meV), as confirmed with several samples. As a commentary on the problems of mobility analysis, one may note that, with similar measurement techniques, Baron et al.<sup>15</sup> found no marked anomaly for As in silicon. Their results (for T = 30 K) correspond to  $E^* \simeq 70$ meV for As, 80 meV for Ga, and 170 meV for In. These track well with the respective ionization energies of these impurities.

Comparable analyses do not seem to be available for  $\mu_N$  over a wide temperature range for germanium containing shallow impurities. The Debye and Conwell 1954 paper<sup>12</sup> on transport in *n*-type Ge made estimates of  $\mu_N$ , which suggest (in the light of the now known effective-mass parameters) approximate parity with  $\mu_E$ . Cyclotron resonance linewidth has been used to measure neutral scattering rates in Ge at liquid-helium temperatures. Thus, Blagosklonskaya *et al.* have measured  $1/\tau$ for electrons scattered by shallow donors,<sup>7</sup> holes by shallow acceptors,<sup>17</sup> and electrons by shallow acceptors.<sup>7</sup>

The latter process may be compared with positron-hydrogen scattering and was found<sup>7</sup> to be a decade weaker than the "Erginsoy" type of process. Otsuka *et al.*<sup>18, 19</sup> used cyclotron resonance linewidth to deduce  $1/\tau$  for germanium containing neutral *multivalent* impurities: divalent Zn and Ni, and trivalent Cu and Au. Here again, the scattering of *electrons* by acceptors was studied. This was up to 50 times weaker than the Erginsoy rate for Zn and Ni. Otsuka *et al.* considered this reasonable, by analogy with the effectiveness of the corresponding atomic process of positron-helium scattering.<sup>2</sup>

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In a study relevant to the present work, Norton and Levinstein<sup>13</sup> analyzed hole transport in *p*-type copper-doped germanium over the range 4–150 K. Copper is trivalent, with a first ionization energy  $E_{a1} = 42$  meV, and could reasonable be expected to be less efficient in neutral scattering than a Group III monovalent acceptor. Thus one might expect to find (as Norton and Levinstein did) that  $A = \mu_N/\mu_E = E^*/E_{em} > 1$ .

The Norton and Levinstein analysis permitted A to be computer-selectable for each sample, but did not allow for temperature dependence of A. Values for A between four and six (i.e.,  $E^*$  from 40 to 60 meV) were found for the various samples. The neutral scattering rate appeared not to increase linearly with the neutral copper density  $N_N$ . That could indicate a scattering contribution by neutral acceptor pairs<sup>20</sup> in the more heavily doped samples.

## IV. MEASUREMENTS WITH MERCURY-DOPED GERMANIUM

Samples used for this work were obtained from apparently weakly compensated sections of Bridgman-grown Ge:Hg crystals. The primary measurements envisaged were of conductivity  $\sigma$  $= ep_0\mu_p$  and of Hall coefficient  $R_H = r_H/ep_0$ . Some samples were bridge-shaped, and others of Maltese-cross shape for van der Pauw measurements.<sup>21</sup> Dc measurements were made with the usual reversals of current and field directions, using an active constant current source and an electrometer readout. Figure 1 shows Hall data so obtained for van der Pauw samples 1, 2, and 3; and for bridge-shaped sample 4.

It was planned that Hall measurements satisfy the high-field criterion (for  $r_H = 1$ ) where possible, and that could always be satisfied for T < 125K. Corrections were applied<sup>22</sup> for a Hall factor  $r_H \neq 1$  to extend the analyses of  $\mu_p(T)$  and particularly of  $p_0(T)$  towards 200 K. That is why the ordinate scale of Fig. 1 is of  $r_H/eR_H$ . Uncertainty over the accuracy possible for  $r_H$  corrections<sup>22</sup> does not affect the mobility deduced for the range 10 < T < 100 K.

Discussions of  $p_0(T)$  for a *p*-type semiconductor containing  $N_a$  principal acceptors are usually couched in terms appropriate for a situation of partial compensation:  $p_0 = N_{ai} - N_D$ . The parameter  $N_D$  is in actuality an algebraic difference between the concentrations of all donors higher in energy than  $E_F$  and all acceptors shallower than the principal species. How  $p_0$  varies with temperature for  $N_a > N_D > 0$  is well known<sup>23</sup> as the solu-



FIG. 1. Temperature dependence of free hole density, as deduced from Hall effect, for four Ge:Hg samples. Sample 1 was partially compensated, and a fit of the data to Eq. (3) yielded  $N_a = 1.06 \times 10^{15} \text{ cm}^{-3}$ ,  $N_D = 1.4 \times 10^{12}$ cm<sup>-3</sup>, and  $p_1 = 1.5 \times 10^{15} T^{3/2} \exp(-0.0931/kT)$  cm<sup>-3</sup>. The line associated with the data points for sample 1 traces Eq. (3) with those values for the parameters. Samples 2, 3, and 4 (of which samples 2 and 3 came from the same Ge slice) all had  $N_a \simeq 2 \times 10^{15} \text{ cm}^{-3}$ , and parameters for Eq. (3) could be determined for these<sup>25</sup> from data for T > 40 K. Hall voltages (and also conductivity voltages) for lower temperatues were impaired by spatial inhomogeneity of the quantity  $N_D$ , the net excess (if any) of donors over shallow acceptors. Thus, such material was unacceptable for mobility analysis, even though it had a finite dark "conductance" down to 10 K.

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$$p_0 (p_0 + N_D) / (N_a - N_D - p_0) = p_1 = (N_v / \beta) \exp(-E_a / kT)$$
.  
(3)

Positive control of the compensation ratio  $(N_D/N_a)$  is necessary when a material such as Ge:Hg is used for extrinsic photoconductive detection.<sup>24</sup>

Advantages were anticipated in making mobility analyses for slightly undercompensated material, in which shallow acceptors slightly outnumbered donors, to make  $N_D$  a small negative quantity. Then  $p_0$  would fall only as far as  $|N_D|$  on cooling, and there should be a wide temperature range of rather slowly-varying hole density and conductivity, convenient for mobility analysis studies.

The undercompensated samples 2, 3, and 4 of Fig. 1 exemplify material which looked superficially attractive, yet which proved unsuitable, because of spatial inhomogeneity of  $-N_D$ . The idiosyncrasies of both conductivity and Hall voltages for such samples below about 40 K (the reason the ordinate scale of Fig. 1 includes the word "apparent"), will be discussed elsewhere.<sup>9</sup> As a passing indication of the delicate and spatially varying balance between donors and shallow acceptors in such material, it is noted here simply that samples 2 and 3 were cut from adjacent parts of the same slice.

Other undercompensated crystals proved unsuitable for different reasons. Some were undercompensated so far as the mercury level was concerned, but showed the 42-meV activation energy of copper contamination on cooling. Other crystals could not provide meaningful data on  $\mu_N$  since  $-N_D$  was too large, and ionized impurity scattering dominated at low temperatures.

Information about  $\mu_N$  was eventually obtained with material of (positive) partial compensation such as sample 1. Conductivity and Hall effect were usually well behaved for this type of material over the measurable temperature range. The range 35-200 K accounted for an eight decade change of  $p_0$ , and an eleven decade change of  $p_1$ , as deduced by a fit of  $p_0(T)$  to Eq. (3) by a critical numerical procedure.<sup>25</sup> The numbers so derived for sample 1 appear in the caption of Fig. 1.

Figure 2 shows hole mobility data for sample 1, as compared with the lattice-scattering mobility  $\mu_L$  reported by Brown and Bray<sup>26</sup> for *p*-type germanium and with curves indicative of two neutral scattering efficiencies. (These curves are discussed in Sec. V.) The upper temperature limit of 100 K for Fig. 2 should ensure  $r_H = 1$  for Hall measurements with fields of a few kilogauss, so that  $\sigma R_H = \mu_p$ . This was verified for various temperatures by measuring Hall voltage versus magnetic field strength.

The data points of Fig. 2 have actually been displaced upwards by 2% and are of  $GoR_H$  with G = 1.02. This was done to bring experiment and calculation into accord over the widest temperature range and follows the practice of Norton *et al.*<sup>13,16</sup> in using a dimensionless factor G close to unity in the mobility analysis, accounting for errors in sample dimensions or geometric regularity.

The open triangles in Fig. 2 show  $\mu_{p}$  as measured in the dark. That procedure could not be extended much below 35 K—at which point the sample resistance reached 10<sup>10</sup>  $\Omega$ , and doubled or more for each further 1 K of cooling. However,



FIG. 2. Experimental points of mobility data for sample 1, as measured in the dark (open triangles) and with weak extrinsic illumination (filled triangles). All data points have been displaced upwards by 2% in order to bring the calculated magnitude and temperature dependence of mobility into registry with the data over the widest temperature range.  $\mu_L$  shows lattice mobility as reported by Brown and Bray.<sup>26</sup> Dashed curve (a) is calculated for lattice, ionized impurity, and neutral acceptor scattering; the latter at a rate for  $\mu_N N_N = 8.8 \times 10^{20}$  cm<sup>-1</sup> V<sup>-1</sup> s<sup>-1</sup> ( $E^* = 50$  meV). Curve (b) is calculated in like manner for  $\mu_N N_N = 4.4 \times 10^{20}$  cm<sup>-1</sup> V<sup>-1</sup> s<sup>-1</sup> ( $E^* = 25$  meV).

the contribution of  $Hg^0$  to hole scattering becomes most important *below* 35 K.

Mobility measurements for sample 1, and for other partly compensated samples, were thus extended to lower temperatures using weak-illumination photoconductive and photo-Hall techniques. Typically,  $\Delta p$  was maintained in the range 10<sup>8</sup> to  $10^{10}$  cm<sup>-3</sup> for the lower temperatures, using a small flux of 300-K thermal radiation. A cooled In As filter blocked the short-wave part that could have caused any intrinsic excitation. It was expected, and was confirmed, that  $\mu_p$  as measured was independent of  $\Delta p$  provided photoionized Hg<sup>-</sup> were much less numerous than those ionized permanently through compensation.

The solid triangles in Fig. 2 show the photoassisted data points of  $G\sigma R_H = \mu_p$  for sample 1. The photo-assisted data blends with  $\mu_p$  as measured in the dark for T > 35 K. This was important to know, since the rise of  $\mu_p$  on cooling, while monotonic, does show significant changes of slope around 40 K. A comparable inflection region was seen with other samples. These were usually less prominent than in Fig. 2, since most other samples permitted only a lesser role for neutral acceptor scattering.

### V. DISCUSSION

Mobility data, such as that in Fig. 2 for sample 1, were analyzed for the contributions of phonons, ionized impurities, and neutral acceptors to the scattering of heavy and light holes. Parameters established by Brown and Bray<sup>26</sup> were used for the ratio of heavy to light holes, and for the magnitudes and temperature dependence of lattice mobilities in those bands. The dashed curve in Fig. 2 shows  $\mu_r$  as deduced by Brown and Bray for the combination of light and heavy holes and confirmed in the work of Norton and Levinstein.<sup>13</sup> The rates assumed for ionized impurity scattering in the two bands also followed the practice of Brown and Bray,<sup>26</sup> based on the Brooks-Herring procedure,<sup>27</sup> and with the coefficients adjusted to take hole-hole scattering<sup>28</sup> into account.<sup>29</sup>

Curve (a) of Fig. 2 was calculated assuming lattice, and ionized impurity, scattering components as noted above and neutral acceptor scattering at the rate corresponding to  $\mu_N N_N = 8.8$  $\times 10^{20}$  cm<sup>-1</sup> V<sup>-1</sup> s<sup>-1</sup>. In terms of the parameter  $E^*$ of Eq. (2), that corresponds to  $E^* = 50$  meV. Curve (b) was calculated assuming neutral acceptor scattering *twice* as strong:  $E^* = 25$  meV, and  $\mu_N N_N = 4.4 \times 10^{20} \text{ cm}^{-1} \text{ V}^{-1} \text{ s}^{-1}$ . It will be seen that curve (a) provides a pretty good fit to the data for T > 50 K, while curve (b) provides approximately the correct increase of neutral acceptor scattering efficiency for T < 35 K. The figure implies that the effective scattering cross section of a neutral acceptor just about doubles between 50 and 35 K. Data from other samples were supportive of that trend.

As was reported in Secs. II and III, temperature dependence of the neutral scattering cross section is a feature of several models for (monovalent) impurities and has been seen in experiments with other systems. However, the rather narrow temperature range of that twofold cross-section increase as seen in Ge:Hg samples did come as a surprise.

Surprising also was the strength of the Hg<sup>0</sup> scattering at all temperatures. Even for T > 50 K, each Hg<sup>0</sup> appears to be twice as efficient in scattering as would have been the case for  $E^* \simeq E_{al}$ . Thus scaling of  $\mu_N$  in proportion to binding energy, as suggested by Baron and McGill<sup>8</sup> for monovalent centers (and which appears to be roughly true for trivalent copper in germanium<sup>13</sup>), is apparently not valid for Hg in Ge.

The unexpected aspects of the scattering rate and its temperature dependence, as noted in the last two paragraphs, could be the consequences, at least in part, of hole scattering by pairs (or larger multiples) of neutral acceptors, when the hole de Broglie wavelength exceeds the average interacceptor spacing  $(3/4\pi N_a)^{1/3}$ . Honig and Maxwell<sup>20</sup> reported the analogous process of electrons scattered by neutral-donor pairs in silicon. Now the wavelengths  $\lambda_{pL}$ ,  $\lambda_{pH}$  for light and heavy holes in germanium have a Maxwellian broadened distribution about

$$\lambda_{\boldsymbol{b}I} \simeq 3\lambda_{\boldsymbol{b}H} \simeq 6000 T^{-1/2} \text{ Å}. \tag{4}$$

Thus for germanium with  $N_a \simeq 10^{15}$  cm<sup>-3</sup> (typical interacceptor spacing  $D \simeq 600$  Å), then  $\lambda_{pL} > D$  for most of the temperature range 10–100 K. Even for heavy holes,  $\lambda_{pH} \rightarrow D$  by the lower end of that temperature range. This makes it likely that the neutral acceptor scattering deduced in the present work—as exemplified by Fig. 2—includes some component of scattering by neutral pairs. One would have more assurance that the effect originally sought, scattering by isolated Hg<sup>0</sup>, would be seen less ambiguously through measurements on less strongly doped samples. However, neutral impurity scattering is too weak to provide much of an effect with further dilution of the scattering species.

For the Ge:Hg system, as in other semiconductor impurity-host systems, the results are only partially satisfying. That uncertainty is likely to continue, unless chance turns up a system for which neutral impurities just happen to be very efficient scatterers. As a corollary to the work reported here on mercury in germanium, it would be interesting to see how lighter members of Group II fare in that host. Otsuka *et al.*<sup>19</sup> have described how neutral zinc in germanium scatters *electrons*, but the scattering of *holes* by  $Zn^0$ (with a first ionization energy of 30 meV) does not appear to have been reported.

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  <sup>29</sup>Allowance was made as a matter of course for ionized impurity scattering in both heavy-hole and light-hole bands, even though the contributions of that process were arranged to be minimal. Ionized donors and acceptors were estimated as providing some 4% of the hole scattering at 100 K, decreasing to about 2% at 50 K, but increasing to some 11% of the total at 20 K.