Donor and acceptor energies for muonium in GaAs

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Analysis of existing data on muonium in GaAs provides an extensive set of ionization energies and barriers for motion and site transitions. Within an accepted model of muonium behavior in III-V compounds, these results establish the energy relationships among observed muonium centers in GaAs, including the metastable states. This analysis places the (0/+) donor level at 0.17 eV below the conduction band edge and the (-/0)acceptor level at roughly 0.60 eV above the top of the valence band for this very light hydrogen isotope, thus locating the muonium equivalent of the H(+/-) pinning level about 0.21 eV above midgap in GaAs, nearly 0.5 eV higher than predicted.

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I. INTRODUCTION

Theoretical modeling of hydrogen impurities in semiconductors carried out over a period of many years has produced the accepted picture of H behavior in these materials,¹ albeit with rather meager direct experimental evidence for the isolated atomic defect except in Si. The indirect verification of these models is mostly from results on the complexes formed by passivation reactions of mobile hydrogen with other impurities in which the addition of H satisfies various bonding mismatches and removes defect levels from the gap, thus eliminating the defect-related electrical activity.

Isolated hydrogen is typically a negative-*U* center, where $U=E_A(-/0)-E_D(0/+)$ is the separation between donor and acceptor thermodynamic levels. Consequently, in most semiconductors, hydrogen shows compensating properties, existing as H⁺ in *p*-type and H⁻ in *n*-type materials under equilibrium conditions.

Isolated hydrogen can exist in three charge states, H^+ , H^0 , and H⁻, with the ionic centers residing at different interstitial sites. For predominantly covalent cubic materials, H⁺ is stable at or near the center of a stretched bond² [bondcentered (BC) site] and H^{-} is stable in the large tetrahedral void³ (T site) of the diamond or zinc-blende structure. Both of these sites can support a neutral center,¹ but the electronic wave functions and the hyperfine interactions are vastly different at the two locations. When H⁰ resides in the void, it is simply an interstitial impurity atom with a large isotropic hyperfine constant, somewhat reduced from the free-atom value by relatively small overlaps with near neighbors. However, in forming H_{BC}^0 , hydrogen has reacted with the host to form a radical in which the unpaired electron resides in antibonding orbitals on its two nearest neighbors,⁴ producing a small anisotropic hyperfine interaction.

Most of the direct experimental evidence supporting the above picture comes from studies of muonium (Mu), a light isotope of hydrogen in which the proton has been replaced by a positive muon. Mu is formed by implanting muons into the semiconductor and nearly always exists as an isolated impurity due to the short muon lifetime $\tau_{\mu} \approx 2.2 \ \mu s$. The experimental time window also means that the mixture of Mu states is often far from equilibrium. Various metastable muonium centers formed during implantation and thermalization can be long lived compared to τ_{μ} and thus experimentally accessible. Previous results for Mu in GaAs identified two Mu⁰ centers,⁵ yielded detailed structures^{2,3,6} for the lowest-energy sites, Mu⁰_{BC}, Mu⁺_{BC}, and Mu⁻_T, and characterized the motion^{7–9} of each state including Mu⁰_T.

For a negative-U impurity such as hydrogen, the (0/+) thermodynamic donor level lies above the (-/0) acceptor level, reversed from the normal ordering. In this case, the neutral state is never dominant and the equilibrium charge-state changes directly from positive to negative when the Fermi level E_F crosses the midpoint between the (-/0) and (0/+) levels, labeled (+/-). Theory predicts¹⁰ that H(+/-) coincides with a more general *charge-neutrality level* and is pinned at zero on the electrochemical scale independent of the host material. A great deal of effort has been directed toward determining the energies for hydrogen donor and acceptor levels in various materials to test these predictions.

Here, we report on results from a large body of experimental work on muonium in GaAs. Much of the basic data are already in the literature but were not previously interpreted to obtain the Mu defect levels. We demonstrate that the Mu isotope of H does indeed have negative-*U* properties in GaAs and establish energies for the Mu defect levels to a reasonable accuracy. We show that donor and acceptor levels are already inverted for the unreacted Mu atom and ions located at T sites in GaAs and measure the Coulomb stabilization for the Mu_T ions. We discuss an interpretation of the motional properties of Mu⁺ centers¹¹ that ties together separate ionization measurements for BC and T sites to establish both the Mu donor and acceptor thermodynamic levels, and thus the pinning energy, relative to the GaAs band edges.

Since the BC site is the most stable location^{1,2,6} for both Mu^0 and Mu^+ with a negligible change in lattice relaxation,

the Mu_{BC}^0 ionization energy provides a direct measurement of the position of the donor level below the conduction band minimum E_C . Determination of the acceptor energy is not as clean. Our measurements directly yield a T-site ionization energy that must be adjusted for the BC to T-site energy difference for Mu^0 to define the proper Mu(-/0) acceptor level.

A direct comparison of Mu versus H is available for silicon, where the above relationships also hold. The muonium results¹² and DLTS measurements¹³ on proton implanted p^+n Si diodes directly give the donor levels as $E_C - 0.21 \pm 0.01$ eV for Mu(0/+) compared to $E_C - 0.175 \pm 0.005$ eV for H(0/+). A larger increase in zeropoint vibrational energy in going from Mu_{BC}^0 to Mu_{BC}^+ compared to that from H_{BC}^0 to H_{BC}^+ can account for this difference. These zero-point energies can be several tenths of an eV for Mu.¹⁴ Both experiments were interpreted^{15,16} as also probing $T^- \rightarrow T^0 + e_c$, yielding $E_{T(-/0)}$ of 0.56 ± 0.06 eV for Mu and 0.65 ± 0.10 eV for H relative to E_C as the most direct acceptor-related measurement. This offset in the T-site acceptor energy can likely be attributed to different zero-point energies for Mu_T^0 versus H_T^0 . The adjustments^{16,17} to obtain the proper (-10) acceptor levels in Si were estimated to be $\sim 0.15 \text{ eV} (\sim 0.17 \text{ eV})$ for H (Mu). The resulting acceptor energies imply that |U| is about 0.15 eV smaller for Mu and place the (+/-) pinning energies in silicon at 0.34 eV (0.30 eV) below E_C for H (Mu), with uncertainties at least 50% larger than the difference.

II. EXPERIMENTAL RESULTS

The data on Mu in GaAs come from several variations of muon spin research methods,¹⁸ or μ SR, in which 100% spinpolarized positive muons are implanted into the sample and the asymmetry of positron emission during muon decay is monitored. Due to parity violation, the positrons are emitted preferentially along the instantaneous direction of the muon spin. For either Mu⁺ or Mu⁻, the spin precesses at the muon's Larmor frequency (135.54 MHz/T) when an external magnetic field is applied perpendicular to the initial polarization. The linewidth, or relaxation rate, for this *diamagnetic muonium* signal decreases as the muon hops more rapidly among equivalent sites. The low-temperature linewidths are nearly identical in GaAs; however, Mu⁺ and Mu⁻ become mobile at approximately 200 and 550 K, respectively,^{8,9,19} so that between these temperatures the relaxation rate identifies which ionic state is observed. Neutral muonium is paramagnetic and precession frequencies are determined by the hyperfine interaction in addition to any applied field. Because Mu_{BC}^{U} and Mu_T^0 have different hyperfine interactions, observed frequencies⁵ definitively identify the Mu⁰ centers.

The above techniques require phase coherence in the spin precession; thus, only states formed within a small fraction of the precession period following implantation can be seen. Another technique, rf- μ SR, in which the magnetic field is applied parallel to the initial polarization and the precession is driven by radio-frequency fields, also detects slowly formed centers and is especially useful in identifying transition final states. Without the rf excitation, precession is not



FIG. 1. Relaxation rates for the two Mu_{BC}^0 spin precession signals in GaAs with $\mathbf{B} || \langle 100 \rangle$. The increase above 110 K directly measures the Mu_{BC}^0 to Mu_{BC}^+ ionization rate and establishes the Mu(0/+) donor level relative to E_C .

normally observed and the depolarization rates provide a signature for various dynamic processes. In particular, when the Mu charge-state fluctuates due to rapid charge exchange, the field dependence of depolarization rates at each temperature yields the hyperfine constant for the Mu⁰ center involved and the rates for both transitions in the cycle.²⁰

As already discussed, the $BC^0 \rightarrow BC^+ + e_c$ ionization energy directly locates the (0/+) donor level. Figure 1 shows the increase in relaxation rate with temperature for the transverse-field spin-precession signals of Mu_{BC}^0 from one of the four samples used to obtain the Mu donor energy in GaAs. The measured relaxation rate is essentially the sum of an intrinsic linewidth for that hyperfine signal plus the ionization rate. The average Mu_{BC}^0 ionization energy from Arrhenius fits places the Mu(0/+) donor level in GaAs at $E_C-173\pm5$ meV, based on a total of ten hyperfine signals from the four samples. Statistical uncertainties for individual values were typically smaller than the spread; thus, we quote the standard deviation. For comparison, DLTS on proton implanted GaAs diodes²¹ puts the H donor level at E_C-141 meV, giving about the same shift for H to Mu as seen for Si.

In GaAs, the T site with Ga nearest neighbors is the muonium acceptor site since it is the stable location for Mu⁻. However, locating the acceptor level requires knowing the energy difference for Mu⁰ in T versus BC sites. It is further complicated by the fact that Mu⁰_T is mobile at all temperatures and the existence of several mechanisms for transitions out of Mu⁰_T. In principle, one can assign the Mu(-/0) acceptor energy based on the energy of Mu⁰_T relative to Mu⁰_{BC} and the activation energy for Mu⁰_T to capture an electron out of the valence band, alternatively viewed as *hole ionization*, T⁰ \rightarrow T⁻+ h_{p}^{+} .

Temperature-dependent diamagnetic rf- μ SR amplitudes from which we extracted the T-site h^+ ionization energy $E_{T(0/-)}$ are shown in Fig. 2. The increase between 300 and 400 K represents growth of Mu_T from an as-implanted Mu_T⁰ fraction. This step is observed in multiple semi-insulating samples and in high-quality liquid phase epitaxial (lpe) layers of very lightly doped *n*-type GaAs,²² all yielding a simi-



FIG. 2. Amplitudes for the diamagnetic rf- μ SR signal from an lpe layer of *n*-type GaAs (filled symbols) plus data including the semi-insulating GaAs substrate (open symbols). The increase near 350 K is assigned to h^+ ionization of Mu⁰_T; the decrease above 450 K is the onset of cyclic charge exchange.

lar energy. Over the same range, two components are seen in the diamagnetic spin-precession signal. The identification of Mu^- as the final state is made by a growing spin-precession amplitude with a relaxation rate consistent with Mu^- , as opposed to a constant-amplitude weakly relaxing component assigned to the mobile Mu^+ centers seen below 300 K. The decrease above 450 K also takes place in two steps, consistent with the initial Mu_{BC}^0 and Mu_T^0 fractions, further implying that the diamagnetic state is different for muons initially occupying the two sites.

We assign 552 ± 38 meV to $E_{T(0/-)}$ based on an average value from five rf- μ SR data sets on different samples. The listed uncertainty is again the standard deviation. This energy defines a T-site acceptor energy relative to the valence band maximum E_V but must still be adjusted for the Mu⁰ site metastability to obtain the correct energy for the Mu(-/0) acceptor level.

Unfortunately, no direct measurement exists for the energy difference between the two Mu^0 states in GaAs. However, we have experimentally defined the relative energies for the two sites by a series of activation energies associated with transitions and motion of the Mu^+ states. Charge transfer in the Ga–As bond, which makes T_{Ga} the stable location for Mu^- , also implies that T_{As} should attract Mu^+ and provide a metastable location.

Longitudinal depolarization rates provide evidence for several different Mu charge cycles in GaAs. The relevant one for our case is active in semi-insulating and lightly doped *n*-type material and fits best as $T^0 \leftrightarrow T^+ + e_c$. Figure 3 shows transition rates into and out of Mu_T^0 for this cycle from two samples. The data for the 10^{15} cm⁻³ *n*-type sample clearly identify the $R_{+/0}$ process as e^- capture, 20 and the two samples yield essentially the same energy for $R_{0/+}$, implying e^- ionization of Mu_T^0 leaving Mu^+ at T_{As} . Thus, the average ionization energy of 453 ± 12 meV measures the depth below the conduction band edge for a metastable T-site donor.

We have thus defined both a donor and an acceptor level for muonium at the T sites in GaAs, with Mu⁺ (metastable) at T_{As} , Mu⁻ (stable) at T_{Ga} , and a mobile Mu⁰_T (metastable) that hops rapidly among both T sites. The h^+ and e^- ionization



FIG. 3. Transition rates for a 0/+ change cycle for Mu_T in semi-insulating (squares) and 10^{15} cm⁻³ *n*-type (circles) GaAs imply a metastable T-site donor level at E_C -0.45 eV.

energies we find for Mu_T^0 imply that Mu_T would constitute a negative-U system even without the reaction to form the Mu_{BC} centers, yielding $U_T \cong -0.36$ eV near 450 K where the gap energy is about 1.35 eV. The Mu_T system in GaAs would be pinned at a T(+/-) level about 0.05 eV above midgap. The decrease in Mu^- amplitude above 520 K, shown in Fig. 2, gives an energy of 1.02 ± 0.07 eV and is assigned to $T^- \rightarrow T^0 + e_c$. Coupled with the h^+ ionization energy for Mu_T^0 , this implies a Coulomb energy of ~ 0.22 eV helping to stabilize Mu^- at T_{Ga} . Geometry and bond ionicity suggest a similar value for Mu^+ at T_{As} .

The final step required to define the proper Mu(-/0) acceptor level is to obtain the T-site energies with respect to those for the BC states. We use data on Mu^+ motion¹¹ to accomplish this with a couple of crucial assumptions. The depolarization function in zero applied field is the most sensitive probe of motion for Mu^+ . The onset of diffusion occurs near 175 K, but the data^{8,11} imply a change in that motion near 220 K.

Figure 4 shows the results of an analysis in which a kink in slope of hop rate versus temperature⁸ is assigned to a change from a diffusing Mu_{BC}^+ state to a higher-energy state where Mu⁺ moves among T_{As} sites. The data are then fitted to separate functions for the two mobile Mu⁺ states. The constant hop rate in Fig. 4(a) for Mu_{BC}^+ at low temperatures represents tunneling among the four BC sites associated with one host atom to which Mu⁺ remains bound.¹¹ For this *p*-type GaAs sample, we obtain $E_1 = 225 \pm 16$ meV as the activation energy for the jump to a different central atom, initiating Mu_{BC}^+ diffusion. Within this model, Fig. 4(b) shows the transition from BC to TAs as reflected in the signal amplitudes, requiring an additional energy of $E_0 = 369 \pm 19$ meV obtained from the displayed fit. Hop rates for motion among the T_{As} sites have an activation energy of $E_2 = 271 \pm 36$ meV as also shown in Fig. 4(a).

In order to complete the determination of the difference in BC- and T-site energies for Mu⁺, we have made one additional assumption that the saddle point for diffusion through T sites is identical to that for the BC-to-T transition. With this assumption, the Mu⁺_T energy with respect to Mu⁺_{BC} is defined as $\Delta_{+}=E_1+E_0-E_2$, which yields $\Delta_{+}=323\pm43$ meV from the results in Fig. 4.



FIG. 4. (a) Hop rates and (b) amplitudes associated with two diffusion paths for Mu^+ motion in GaAs yield barriers for Mu^+ motion and a BC-to-T_{As} site change.

Finally, having obtained the difference in energy between the T and BC sites for Mu⁺, the difference for Mu⁰ is then $\Delta_0 = \Delta_+ + E_{BC(0/+)} - E_{T(0/+)}$, which implies that Mu⁰_T is only slightly metastable with respect to Mu⁰_{BC}, giving $\Delta_0 = 43 \pm 45$ meV. We can now adjust the thermodynamic acceptor level to lie at $E_{T(0/-)} + \Delta_0$ above the valence band edge, yielding a Mu(-/0) acceptor energy of $E_V + 595 \pm 62$ meV in the 350-500 K range. Figure 5 displays the final experimental energy diagram for the Mu states observed in GaAs as a function of Fermi energy, including the lowest-energy sites for all three charge states and the metastable T-site locations for mobile Mu⁰ and Mu⁺ centers. The energy relationships



FIG. 5. Experimental formation energies as a function of E_F for all of the muonium charge states and sites observed in GaAs. Bold arrows indicate the features for which the energies are most directly determined as discussed in the text.



FIG. 6. Comparison of the Mu defect levels obtained in Si, Ge, and GaAs. Symbols represent the measured BC donor energy and the T-site acceptor energy, while the bars include the inferred or approximated adjustment of acceptor levels to account for Mu^0 site metastability.

inherent to this diagram have been used throughout the preceding analysis.

III. DISCUSSION AND CONCLUSIONS

The analysis we report in the previous section places the muonium donor and acceptor levels in GaAs at $E_C - 173 \pm 5$ meV and $E_V + 595 \pm 62$ meV, respectively. These results then place the Mu(+/-) pinning level at 211±33 meV above midgap. Using a gap of 1.35 eV at 450 K, the energies we obtain also yield $U \cong -0.57$ eV for Mu in GaAs.

Placement of the Mu(+/-) level above midgap in GaAs is fully consistent with our general observations that the dominant muonium state is Mu⁻ for *n*-type concentrations above 10^{17} cm⁻³ but is Mu⁺ at 10^{15} cm⁻³. Charge-cycle results suggest that the switch from Mu⁺ to Mu⁻ as the dominant state occurs in the mid- 10^{16} cm⁻³ range. Within the band alignments of Ref. 10, our placement of Mu(+/-) in GaAs is essentially identical with what we find for Si and Ge,¹⁷ as demonstrated in Fig. 6. This agreement is consistent with the predicted universality, but the resulting Mu(+/-) energy is almost 0.5 eV above the predicted H(+/-) pinning level and well above other placements²³ of the chargeneutrality level in GaAs.

As briefly discussed for silicon, the energies we obtain include the zero-point energies which are the primary origin of any offset between Mu and H defect levels, if one assumes no fundamental differences in the occupied sites. Because the muon mass is only approximately one-ninth that of a proton, the zero-point energy for Mu is roughly three times that for H in any particular state and can be a few tenths of an eV.14 However, it is more specifically differences in zeropoint energy for different charge states and different sites that are important here. In the BC site, the bond is weaker in the neutral state since the e^- is in an antibonding orbital; thus, the vibrational frequency and zero-point energy are larger in the positive charge state. This difference is greater for Mu and shifts the BC⁺ energy line in Fig. 5 upward with respect to BC⁰; thus, the BC donor level is moved downward, lying further away from the conduction band minimum for Mu compared to H. This shift is about 30 meV in both GaAs and Si based on the experimental results, as previously

mentioned. Detailed arguments for the acceptor level are made difficult by rapid tunneling of the Mu_T^0 state and by the complicated potential energy surface within the T-site region,¹ which becomes much more important for H. No data on the H acceptor level exist for GaAs; thus, we rely on the previously discussed results for Si as the best indication of what shift might be expected. There, the T(-/0) level moves upward by nearly 100 meV for Mu compared to H, lying further from the valence band maximum. Only the *difference* in magnitude of these oppositely directed shifts is relevant to the (+/-) level. Based on the comparisons we cite for Si, the net effect should yield a relatively small upward shift of the (+/-) pinning energy for Mu compared to H, on the order of 50–100 meV at most. We conclude that the half an eV offset from the predicted hydrogen pinning energy far exceeds any expected differences related to the much lighter muonium mass.

The results we have presented also establish a T-site donor level at E_C -453 meV. Above roughly 550 K, the 0/+ cycle associated with this level dominates the μ SR data for semi-insulating and weakly *n*-type GaAs samples, with both charge states highly mobile at these temperatures. Ionization of the metastable T⁰ center during this charge cycle was one of the crucial ingredients in determining the Mu^T_T formation energy. In our analysis and in producing Fig. 5, we have used transitions from the T⁰ state to the T⁺ and T⁻ states in defining the T-site donor and acceptor level energies, with an e^- or h^+ going to their respective bands. We defined the extra Coulomb energy related to T⁻ by emission of an e^- to the conduction band and expect a similar Coulomb energy associated with the T⁺ state. If one instead uses the carrier emission transitions from T[±] to yield T⁰ in order to locate the T⁰

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energy with respect to the *occupied* T-site ionic states, then one would have to locate the T⁰ energy 0.22 eV higher than shown in Fig. 5. We note, however, that this placement of T⁰ would not result in any shift in the Mu(+/-) level since the acceptor level adjustment would also increase by the same amount leaving Mu(-/0) also unchanged. This ambiguity in T-site levels will always be present in materials with any ionic content to the bonding, but is not relevant to the BC donor level due to the nature of that site.

In conclusion, we have experimentally placed the Mu donor and acceptor levels in GaAs at E_C -173 meV and E_V +595 meV, respectively. These energies locate the muonium pinning level Mu(+/-) at 211 meV above midgap in GaAs, which places it nearly 0.5 eV above the predicted location¹⁰ of H(+/-) within the calculated band alignments. This is well outside the expected adjustment for larger zeropoint energies for Mu compared to H, which can account for only 10%-20% of this difference. Comparison of these results with earlier data for Mu in Si and Ge provides support for the claim of universal pinning. However, the higher than predicted pinning energy implies that more materials than anticipated may actually be subject to *n*-type doping by unintentional hydrogen impurities.

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