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Donor level of interstitial hydrogen in CdTe

Vl. Kolkovsky,[1,2] V. Kolkovsky,[3] K. Bonde Nielsen,[1] L. Dobaczewski,[3] G. Karczewski,[3] and A. Nylandsted Larsen[1]
[1]*Department of Physics and Astronomy, University of Aarhus, DK 8000 Aarhus C, Denmark*
[2]*Institut fur Angewandte Physik, Technische Universität, 01062 Dresden, Germany*
[3]*Institute of Physics, Polish Academy of Sciences, al. Lotnikow 32-46, 02-668 Warsaw, Poland*
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The first results demonstrating the existence of a donor level of interstitial hydrogen in CdTe are presented. From the Arrhenius analysis, this donor level is characterized by an activation energy for electron emission $E_C - E_t = 0.06$ eV and a pre-exponential factor equal to 2×10^5 s^{-1} K^2, and is observed after a low-temperature implantation with protons. Above 75 K the hydrogen-deep donor state is unstable converting to a more stable configuration which is believed to be a negatively charged form of isolated H. The results are analogous to what has been observed for interstitial hydrogen in GaAs.

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I. INTRODUCTION

Cadmium telluride and related compounds are increasingly important due to their many applications as x-ray or gamma detectors and in thin-film photovoltaic technologies.[1] However, similarly to other semiconductors the properties and, accordingly, the performance of CdTe-based devices depend to a great extent on the type and concentration of mobile impurities such as transition metals (TMs) and hydrogen, which may be unintentionally introduced during processing. Moreover, in many semiconductors hydrogen can interact with TM impurities leading either to their passivation or to the formation of new electrically active TM complexes.[2,3] This can significantly impede the analysis and understanding of a variety of peculiar effects which are linked to the presence of TM defects. Due to the recognition of its increasing importance, the role attributed to hydrogen in CdTe-related materials has attracted an increased interest in the last decades.[4] The fundamental processes of hydrogen incorporation and its interaction in an isolated form with a perfect semiconductor crystal of CdTe are, however, still not known. It has been already evidenced in previous investigations that the most fundamental hydrogen-related defects in group IV semiconductors are interstitial hydrogen atoms occupying the bond-center (BC) site or the interstitial tetrahedral site (T).[5–8] Both configurations are well understood in silicon and germanium based on investigations using various experimental techniques such as Fourier-transform infrared-absorption spectroscopy,[9,10] electron-paramagnetic resonance,[11] deep-level transient spectroscopy (DLTS), and high-resolution Laplace DLTS.[5–8] In these materials the H impurity is found to exist in three different charge states H^+, H^0, and H^- depending on which site in the crystal lattice it occupies. These charge states give rise to either a donor level $(0/+)$ ascribed to hydrogen at the BC site or an acceptor level $(-/0)$ ascribed to the T interstitial site. However, in contrast to Si the single acceptor level of hydrogen in Ge was conjectured to be resonant with the valence band.[8] Another important feature of the interstitial H defect in group IV semiconductors is linked with its negative-U property. In this case the "normal" sequence of levels is reversed and the single donor level is situated closer to the conduction band than the single acceptor level. A similar behavior of hydrogen has been also recently predicted for other III-V and II-VI group of semiconductor compounds.[12]

Recently it was demonstrated that interstitial hydrogen in GaAs introduces a donor state in the band gap with an activation energy for electron emission of 0.14 eV.[13,14] The structure of this defect was interpreted as an unresolved signal from regular bond-center hydrogen H(BC) and bond-center hydrogen H(BC′) which is perturbed by the local strain from a neighbor point defect.[13] While the regular bond-center configuration of H was found to be stable up to 450 K its perturbed configuration is unstable above 120 K where it converts to a more stable acceptor state.[13] Similarly to Si and Ge the negative-U property of interstitial hydrogen was demonstrated for GaAs as well and other properties of interstitial hydrogen in GaAs were found to be analogous to those of interstitial hydrogen in Si and Ge.

As it has been mentioned above, there is no direct experimental information regarding the individual isolated hydrogen states in Cd-related compounds and most reported properties have been inferred indirectly. For example, muonium is known to behave as a light isotope of hydrogen with analog electronic properties and thus, the muon spin-resonance (μSR) technique has been often used for investigations of the properties of isolated hydrogen.[15–18] Gil *et al.*[16] demonstrated that shallow donor muonium states are generic to the II-VI semiconductor compounds CdS, CdSe, and CdTe. Based on observations of these shallow donors, electronic levels of isolated hydrogen were predicted to be located in the upper half of the band gap. These results are consistent with calculations of Van de Walle and Neugebauer[12] in which the existence of the universality of the position of the $(-/+)$ level of isolated hydrogen was shown in various semiconductors. Another important conclusion drawn from the μSR experiments is that, in contrast to the behavior of muonium in other Cd chalcogenides, in n-type CdTe it forms deep donor states as well. These deep states were proposed to arise as a consequence of the cubic symmetry of the CdTe crystal lattice, where the trapped atom in the neutral charge state can be stabilized in the tetrahedral metal-ion cage for which there is no equivalent position in the hexagonal lattice of CdS and CdSe.

In the present work the existence of the donor level of interstitial hydrogen in CdTe will be demonstrated. This electronic level is in the upper half of the band gap very close to the conduction band. A detailed analysis of isolated hydrogen-related defects generated by a low-temperature proton implantation based on *in situ* applications of high-resolution Laplace DLTS (Ref. 19) is presented.

II. EXPERIMENTAL PROCEDURE

The samples were prepared from 5 μm thick, n-type I-doped CdTe layers grown by the molecular-beam epitaxy technique on n^+-GaAs substrates. The carrier concentration in the substrate was about $\sim 10^{18}$ cm^{-3} while that in the CdTe:I layer was found to be 8×10^{14} cm^{-3} at room temperature from capacitance-voltage (CV) measurements performed at 1 MHz and it stayed practically unchanged in the whole temperature range investigated. Prior to deposition of the CdTe:I layer, the GaAs substrate was covered with about 60 Å thick, undoped ZnTe which was used to reduce the strong lattice mismatch between CdTe and GaAs, and to stabilize the growth in the $\langle 100\rangle$ direction. Au/CdTe Schottky diodes were formed immediately after the growth process by deposition of gold (85%) and palladium (15%) using the rf magnetron sputtering method. The samples were mounted on the cold finger of a cryocooler and implanted with protons or helium at temperatures around 30 K. The implantation energy was chosen so that the peak of the implant matched with the depletion width of the diode (around 2.7 μm) under suitable reverse bias conditions at the implantation temperature. For hydrogen this energy was 400 keV and for helium 1.1 MeV. The CdTe samples were implanted with hydrogen with a series of subsequent irradiations up to a total dose of 5×10^{11} cm^{-2} and measurements were performed after each individual irradiation. Finally, the thermal stability of defects was determined from a series of 15-min-isochronal annealing sequences performed in the 40–110 K range. During each step the reverse bias of -5 V was applied to the Schottky diode.

III. EXPERIMENTAL RESULTS

A. Implantation

Figure 1 shows depth profiles of carrier concentrations of a CdTe layer derived from CV measurements before (crosses) and after (triangles) proton implantation to a total dose of 5×10^{11} cm^{-2}. The distance accuracy for the CV measurement is defined by the Debye length for a given carrier concentration and temperature and here it equals to 0.06 μm. A comparison of the CV profiles before and after implantation clearly indicates the presence of a donor generation at a depth of about 2.7 μm caused by the implantation. Moreover, increasing the implantation dose, for the sake of clarity not shown in the figure, leads to the introduction of additional donors. One should emphasize that the whole profile spreads over 1 μm and this together with the maximum of the donor generation observed at about 2.7 μm fits well with the hydrogen-implantation profile as deduced from transport of ions in matter calculations.[20]

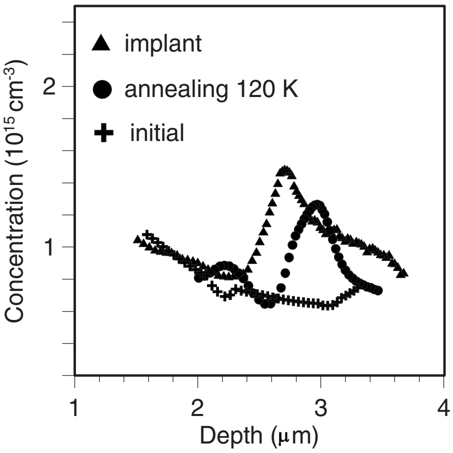

FIG. 1. Carrier concentration versus depth derived from the CV profile before (cross symbols) and after [triangles and round symbols (see details in the text)] implantation. The increase in the carrier profile at around 2.7 μm coincides with the peak concentration of protons.

Figure 2(a) depicts a typical Laplace DLTS spectrum recorded at 44 K for the CdTe sample implanted with protons. The spectrum of Fig. 2(a) was measured with a reverse bias of -5 V and a filling-pulse voltage of -2 V. According to the CV profiling, this corresponds to the depth range of 2.2–2.8 μm below the metal-semiconductor junction. In this case two well-resolved dominant peaks are observed at emission rates of 2 and 40 s^{-1}. The amplitudes of these two lines increase linearly as a function of the proton dose. No signals were observed at this measurement temperature prior to the proton implantation.

In Fig. 2(a) both H60 and H70 peaks were found to be much broader than delta peaks which are expected to appear from nearly monoexponential thermal emission process as a result of the mathematical transient processing used in the Laplace DLTS method (see Ref. 19 for more details). It can be due to the fact that in this case the carrier concentration evaluated from the CV measurements at about 50 K was significantly higher (about tens times) compared with that,

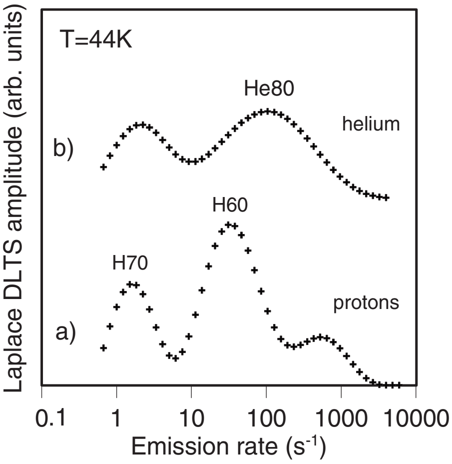

FIG. 2. Laplace DLTS spectra recorded at 44 K in (a) proton and (b) helium-irradiated CdTe.

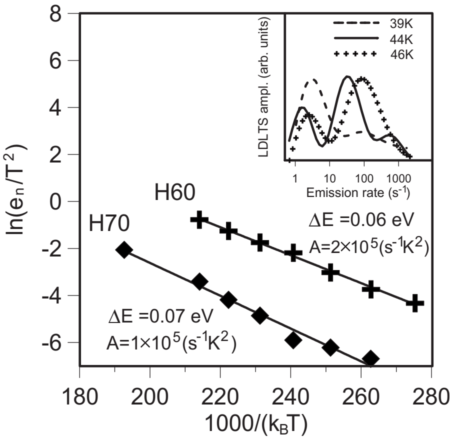

FIG. 3. The Arrhenius plot of the H60 and H70 signals depicted in Fig. 2(a). The inset shows the Laplace DLTS spectra taken in proton-irradiated CdTe at different temperatures.

for example, of GaAs in which narrow deltalike peaks were observed.[13,14] Thus, in order to obtain reasonable amplitudes of the Laplace DLTS signals much larger doses of H were implanted into CdTe. This in turn leads to a higher damage level and more local strain in the crystal resulting in the Laplace DLTS peak broadening. Furthermore, the higher electron concentration also means a larger electric field in the space-charge region, which influences the emission from a donor state in the *n*-type material (see, e.g., Ref. 19). As a result, the Laplace DLTS peaks observed for shallow junctions implanted with hydrogen are rather broad. This is also consistent with the significant broadening of the peak in GaAs samples with a higher dopant concentration of about 10^{16} cm^{-3} for which also higher doses of H implants were used. On the other hand, numerous numerical tests performed on procedures used for the spectra calculation together with measurements performed in similar less advantageous experimental conditions allowed us to conclude that the peak amplitude and average emission rate are still very reliable experimental parameters. Thus, the broadening of the H60 and H70 lines observed in the Laplace DLTS spectra does not influence our conclusions presented below.

As a reference experiment, helium implantations with doses leading to similar damage levels have been performed. In this case two peaks with emission rates of 3 and 100 s^{-1} are observed [Fig. 2(b)]. The absence of the peak at the emission rate of 40 s^{-1} labeled as H60 in Fig. 2(a) shows unambiguously that this emission peak is related to hydrogen. In the following we will mainly concentrate on the properties of this line. As will become apparent the H60 signal observed in the Laplace DLTS spectrum can be correlated with the donor $H^{+/0}$ signal stemming from interstitial hydrogen; this is in analogy with Si (Refs. 5 and 6) and GaAs.[13] From the Arrhenius analysis of the emission process it is found that this level is characterized by an activation enthalpy of electron thermal emission of $E_C - E_t = 0.06$ eV and a pre-exponential factor equal to 2×10^5 s^{-1} K^2 (Fig. 3) where E_C is the conduction-band edge and E_t is the defect-level position. The inset in Fig. 3 shows the Laplace DLTS spectra taken at different temperatures. The dominant peak is broad, what has been discussed previously, and it shifts as a function of temperature. This effect shows that it represents the thermal emission process. Moreover, the points on the Arrhenius diagram are not scattered much around a straight line indicating again that the average emission rate of the peak is a meaningful experimental parameter. The defect concentration corresponding to the H60 peak just after H implantation calculated as the integrated area under the Laplace DLTS peak is $\sim10^{14}$ cm^{-3}. This value is significantly smaller than the peak concentration of the as-implanted donor concentration of $\sim1\times10^{15}$ cm^{-3} introduced at about 2.7 μm and shown in Fig. 1. However, one should remember that the value of about 10^{14} cm^{-3} obtained using the Laplace DLTS method is an average one for the volume of the space-charge region probed by the Laplace DLTS pulses. If the CV implant distribution (a difference between triangles and crosses in Fig. 1) is averaged over the same region then the average value of about 4×10^{14} cm^{-3} is obtained, which is much closer to the value obtained from the Laplace DLTS method. One should also emphasize that in the present case doses used for the H implantation were about 10^2-10^3 times higher than those used in Refs. 5, 6, 13, and 14. Due to this a significantly higher damage level and, accordingly, a higher number of implantation-induced defects such as, for example, vacancy-related defects, which are known to be very effective traps for H species,[2–4] is expected to be introduced in CdTe as well. Following this idea we believe that a part of hydrogen can be trapped by such defects and, therefore, does not contribute to the H60 signal observed just after the low-temperature implantation.

Finally, it is important to note that the introduction of hydrogen into the samples does not lead to the appearance of any other dominant peaks in the conventional DLTS spectra except these two lines observed using the Laplace DLTS technique.

B. Annealing studies

The normalized changes in the H60 trap concentration upon 15-min-isochronal annealing steps, where bias has been applied to the sample, are shown in Fig. 4. Similarly to the case of GaAs no significant differences between the bias-off and bias-on annealing behavior have been observed. For a comparison the results of the He80 defect annealing, which is observed after helium implantation, are also in this figure presented. As seen in Fig. 4 the results of the H60 and He80 annealing differ substantially. While after the H implantation the intensity of H60 increases to almost 300% of its initial value at about 80 K, and in the range of 90–110 K the H60 line anneals completely, the He80 signal disappears at 80 K. This behavior clearly indicates that these two signals originate from different defects.

C. H60 Laplace DLTS signal recovery effect

The important characteristic feature of the H60 trap is that after the annealing process below 120 K for 15 min the intensity of the Laplace DLTS signal corresponding to this defect can be completely recovered to its initial value by subsequent white light illumination at about 45 K. Moreover, the procedure of recovery of H60 can be cycled with no loss

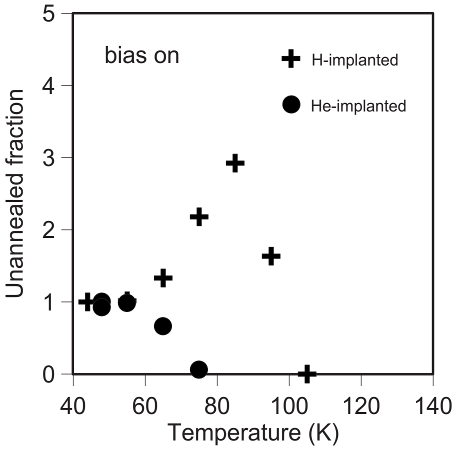

FIG. 4. The normalized change in concentration of the traps responsible for the dominant lines observed after hydrogen and helium implantation in the Laplace DLTS spectra upon 15-min-isochronal annealing steps (see details in the text). During each step the samples were biased with −5 V.

or gain of the total signal. The regeneration of the H60 signal strongly suggests that this trap can be converted to and from a configuration unobserved by the Laplace DLTS technique. As mentioned previously, if the H60 trap is the single donor state of isolated hydrogen then we anticipate the existence of an acceptor level associated with hydrogen occupying the T site in the crystal lattice of CdTe. However, there is no obvious indication for the existence of such level in the conventional DLTS spectra recorded after the implantation. On the other hand, if the negative-U ordering of the isolated hydrogen electronic levels in CdTe applies then the acceptor level can be deeper in the band gap and presumably nonaccessible for the DLTS measurements for the used samples.

The capacitance-voltage measurements performed after annealing at 120 K and subsequent illumination provide further support to our interpretation. As seen in Fig. 1 (circle symbols) such an annealing leads to a decrease in the total number of electrically active donors introduced by the implantation process. This reduction can be also interpreted as the appearance or introduction in this region of some number of acceptors. However, a subsequent illumination shifts the compensation profile back to its initial position (shown with triangles in Fig. 1). This effect is similar to the conversion observed for the H60 Laplace DLTS signal and this process can be repeated without any loss of the total signal. Additionally, considering the spatial overlap of the hydrogen implants and the implantation generated vacancies; it should be emphasized that vacancy-related defects (and hydrogen-related defects other than H60) may contribute to the implantation-induced donor generation seen in Fig. 1. As a consequence the broad peak structure observed at about 3 μm in the depth profile in the nonannealed sample is believed to be due to the contribution of vacancy-related defects created just after implantation some of which could possibly be disturbed by the presence of hydrogen. However, we firmly believe that these defects are not related to the H60 peak due to their different annealing behavior and that they are not influenced by the white light illumination.

IV. DISCUSSION

After annealing and subsequent illumination the concentration of H60 calculated from the amplitude of the Laplace DLTS peak is found to be $\sim 3 \times 10^{14}$ cm^{-3}. Due to the similar dynamic behavior observed for the significant part of the donors generated by implantation (the main part of the recovered signal in the CV profile) we link these donors with the hydrogen-related defect observed with the Laplace DLTS method. The concentration obtained from the Laplace DLTS spectra is less than the one from the CV profiling but this discrepancy can be essentially explained taking into account the satellite peak revealed in Fig. 2(a). Until now we have focused our attention on the dominant H60 line observed in the spectrum. However, the other peak observed at an emission rate of 2 s^{-1} could be also hydrogen related. From the Arrhenius analysis of the emission process this defect was found to be characterized by an activation enthalpy of electron emission of $E_C - E_t = 0.07$ eV and a pre-exponential factor equal to 1×10^5 s^{-1} K^2 (see Fig. 3). This satellite of the H60 line behaves qualitatively almost identical to the H60 defect during 15-min-isochronal annealing steps. A similar amplitude increase was observed at around 80 K and this signal together with the H60 line disappears at around 100 K. This is different from what is observed for the signal at a similar emission rate after helium implantation. Thus, we can argue that both the signal at 2 s^{-1} and the H60 lines might be stemming from the interstitial hydrogen. Moreover, as shown in Fig. 1 the annealing of these defects and their recovery with a white light illumination is apparently linked with generation of acceptor and donors centers, correspondingly. These processes can be cycled with no loss or gain of the total signal. Therefore, in analogy to similar properties of isolated hydrogen in Si, Ge, and GaAs it is very likely that both of these signals originate from the $H^{+/0}$ donor signal of isolated hydrogen. Following this idea we might assume that both of them have the same structure but that one of the defects is perturbed by the presence of some crystal imperfection. This stable hydrogen position is in the case of Si related to the presence of interstitial oxygen[5,6] while in GaAs the EL2 defect or oxygen is believed to play the stabilizing role.[13] Our experimental techniques do not allow us to determine unambiguously the cause of this perturbation in CdTe. However, it should be noticed that in contrast to the case of Si and GaAs, where the perturbed and unperturbed hydrogen center have identical electronic levels, the perturbation acts differently for CdTe. Here the neutral and positive charge states are affected differently as deduced from the observed shift in level position. On the other hand, the annealing properties are more similar indicating that barriers are less influenced in this case.

From previous μSR measurements the positive ion Mu$^+$ is known to be trapped either at the bond center in covalently bonded materials (Si, Ge, GaAs, etc., where it is attracted to the high density of spin-paired bonding electrons) or at the antibonding site, close to the anion, in more ionically bonded materials (CdS, CdTe, ZnO, GaN, etc.). On the other hand, the negative ion Mu$^-$ is nonbonding and sits at an open interstitial site at the tetrahedral interstitial site in cubic materials or at the corresponding site in hexagonal crystals.[14] Due to similar properties isolated hydrogen is expected to behave in the same way as well. In the present case the diode was unbiased during the implantation. Then the implanted hydrogen captures electrons and thus, when the temperature is high enough it begins to migrate in the neutral charge state. Comparing with previous studies in other semiconductors (Si, Ge, GaAs, and Cd chalcogenides)[5–8,13,16–18] we can assume that as-implanted hydrogen in CdTe may end up either in an antibonding position as metastable H^0 or in a near-T site as H^- upon capture of an additional electron. This picture anticipates a negative-U character and the existence of a barrier separating the $H^0(AB)$ and $H^0(T)/H^-(T)$ configurations. Then, following this idea it is very likely that the recovery of the H60 signal by illumination may be interpreted as a direct conversion from the $H^-(T)$ state to the $H^+(AB)$ state in the present case. The conversion process of the $H^-(T)$ state can be a photoexcitation to the conduction band of electrons trapped by the defect if photons with energies below the band gap are involved. The above band-gap photons create in CdTe minority holes which can very effectively recombine with $H^-(T)$ in the acceptor state leading to the hydrogen ionization as well.

The presence of the donor hydrogen-related electronic levels close to the conduction band is consistent with predictions by Van de Walle and Neugebauer.[12] As mentioned in the introduction, using *ab initio* calculations they showed that the position of the $(-/+)$ reference level of isolated hydrogen on an absolute energy scale, with an uncertainty corresponding to the accuracy of the conduction-band offsets in CdTe, should be located in the band gap similar to the one in GaAs. From our previous works[5–8,13] the position of the donor level of interstitial hydrogen was found to be constant within the experimental error on the same absolute scale for Si, Ge, and GaAs (including the case of InP where the donor level has not been observed)[14] despite the fact that such an alignment for the donor level has not been foreseen by the theory of Ref. 12. On the other hand, taking into account these phenomenological observations one might expect that the donor level of isolated hydrogen in CdTe should be located close to the conduction band as it is observed for GaAs as a result of similar band offsets calculated for these materials and this is indeed observed in the present work.

Additionally, the identification in the present work of the $H^{+/0}$ signal as $H(AB)$ is in accordance with the conclusions obtained using the μSR studies. As mentioned above, Gil *et al.*[16] concluded that there is the coexistence of deep and shallow states of isolated hydrogen in CdTe. This behavior is fully consistent with a negative-U character of hydrogen, if the $(0/+)$ donor level is close to the conduction band and the $(-/0)$ acceptor level remains deep in the gap. Within the experimental error we can correlate the dominant signals in the Laplace DLTS spectra in the present work with deeper donor levels predicted by the μSR studies. On the other hand, the presence of the negatively charge state of hydrogen observed during the annealing and subsequent illumination experiments could be linked with its single acceptor state. Within the present experiments one cannot definitely conclude the level position of this defect in the band gap. However, due to the experimental analogy to GaAs this level can be foreseen to be located in the lower half of the band gap of CdTe.

V. CONCLUSION

In conclusion, the donor level of isolated hydrogen has been observed in n-type CdTe using high-resolution Laplace DLTS studies. From the Arrhenius analysis this donor level is characterized by an activation energy for electron emission of $E_C - E_t = 0.06$ eV and a pre-exponential factor equal to 2×10^5 s^{-1} K^2. By analogy to other semiconductors the slightly perturbed form of isolated hydrogen with an activation energy for electron emission of $E_C - E_t = 0.07$ eV and a pre-exponential factor equal to 1×10^5 s^{-1} K^2 was proposed to exist in CdTe as well. These results were found to be consistent with both theoretical predictions and the μSR studies.

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