

Positron annihilation at vacancies in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ crystals after electron irradiation

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Acceptors and vacancy-type defects have been investigated in traveling-heater-method-grown $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ crystals by measuring the positron lifetime after 2.5-MeV electron irradiation in liquid hydrogen and annealing from 77 to 340 K. The results give evidence that negative V_{Hg} vacancies are introduced. They disappear slowly at room temperature over several weeks. Another monovacancy is also observed when the Fermi level is high in the gap. It is tentatively identified with the V_{Te} vacancy. A recovery stage is observed between 150 and 280 K, the nature of which is discussed.

I. INTRODUCTION

For more than three decades the narrow-gap semiconducting crystals of $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ (MCT) have been studied.^{1,2} Although MCT has many problems due to lattice, surface, and interface instabilities,³ it is still especially useful as a detector material for 8–14 μm atmospheric transmission windows. Of particular interest is the nature of intrinsic point defects, which are electrically active and influence the electronic properties significantly. The major problems under discussion are the successful p_+ to p_- or n -type conductivity conversion. It is produced by appropriate annealing or ion irradiation and the problems are focused on the nature of point defects playing an important role in the conversion.

Mercury vacancies (V_{Hg}) are thought to be responsible for the p_+ -type behavior in bulk materials grown by the traveling-heater method (THM) from the Te-rich side of the phase diagram.⁴ Annealings under mercury-saturated conditions lead to p_- or n -type materials depending on the residual impurity concentration.

Electron irradiation as well as ion implantation converts as-grown p_+ -type materials to n -type materials. The process is so efficient that ion implantation is currently used to prepare n - to p -type junctions in the device industry.^{5–10} Irradiation introduces different types of defects and several models have been proposed to describe the process underlying the conversion. The main ideas which have been discussed are the following: (i) interstitial atoms annihilate with the acceptor-type vacancies and reduce their concentration, (ii) the native acceptor defects (V_{Hg} , residual impurities) are overcompensated by irradiation-induced donors.

To get information about the defects in MCT, several methods have been used: Hall effect and conductivity characterization, deep-level transient spectroscopy, photoluminescence, and electron paramagnetic resonance.

So far results on electron-irradiated effects in MCT systems have mainly been investigated by Hall effect measurements.^{11–14} To understand better the role of vacancies in the p -to n -type conversion we use here positron annihilation. It has indeed the unique advantage to selectively detect vacancy-type defects. It has provided direct evidence that p -type as-grown crystals contain negative vacancies.^{15,16} It has also been successfully applied to the characterization of vacancies in electron-irradiated Si (Refs. 17–20) and GaAs.^{21–23} However, there are comparatively many fewer studies in II-VI compounds where only in ZnS and ZnSe (Ref. 24) have electron-irradiation-induced vacancies been studied by positron annihilation techniques.

In this work, we show that positron lifetimes give evidence that two types of monovacancies are introduced by 2.5 MeV electron irradiation in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$. One can be identified with negative V_{Hg} vacancies and the other one is tentatively identified with neutral V_{Te} vacancies. At low temperature, positron trapping at V_{Te} vacancies dominates for annealing from 77 to 350 K whereas V_{Hg} vacancies are detected only after aging at room temperature.

II. MCT SAMPLES AND IRRADIATION DETAILS

Samples of $5 \times 5 \times 1 \text{ mm}^3$ were cut from $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ wafers provided by la Société Anonyme des Télécommunications (France) (SAT) which have undergone stoichiometric annealing at a temperature of 260 °C for 15 days. Such annealings remove the vacancies which are observed in as-grown p -type samples.^{15,16} Irradiation with 2.5 MeV electrons was performed at a temperature of 20 K in the liquid-hydrogen cryostat of the Van der Graaf accelerator at the Laboratoire des Solides Irradiés at Ecole Polytechnique. Two pairs, MCT1 and MCT2, were irradiated at the fluences of 5×10^{18} and 10^{18}

TABLE I. Electron concentrations determined from Hall effect measurements performed before and after 2.5 MeV electron irradiation at 20 K and aging for 6 months at room temperature. The third column gives the electron concentration expected at 20 K on the basis of results previously obtained by Favre, Konczykowski, and Blanchard (Ref. 13).

Crystal and fluence ($e^- \text{cm}^{-2}$)	Before $n_{77 \text{ K}}$ (cm^{-3}),	After $n_{20 \text{ K}}$ (cm^{-3}), calc.	After aging $n_{77 \text{ K}}$ (cm^{-3})
MCT1 5×10^{18}	3×10^{15}	1.3×10^{19}	6×10^{16}
MCT2 10^{18}	3×10^{15}	4×10^{18}	4×10^{16}

$e^- \text{cm}^{-2}$, respectively (Table I). Each of them was irradiated on both sides.

Annealing up to 77 K took place during the transfer and the storage of the samples in liquid nitrogen. The positron ^{22}Na source was sandwiched between each pair into the sample holder of the lifetime spectrometer under liquid nitrogen. Isochronal annealings of 30 min were performed *in situ* from 77 to 330 K by steps of 20 K. The lifetime spectra were recorded as a function of annealing or measurement temperature by using fast-fast coincidence spectrometers²⁵ with a full width at half maximum resolution of 270 ps. A high number of 2×10^6 annihilation events was accumulated in the spectra in order to reliably perform the decomposition of the spectra into a sum of exponential components.

The source contribution is constant with temperature and corresponds to about 7%. After source and background corrections, one or two exponential decay components were used to fit the lifetime spectra $n(t)/n_0 = \sum_i I_i \exp(-t/\tau_i^*)$, where I_i ($\sum_i I_i = 1$) is the relative intensity of the lifetime τ_i^* . The average positron lifetime τ coincides with the center of mass of the lifetime distribution and is calculated from the decomposition of the spectra as $\tau = \sum_i I_i \tau_i^*$.

Hall effect measurements were carried out before irradiation and after aging at 300 K for 6 months. As earlier observed,¹⁴ irradiation and aging result in a strong increase of the electron concentrations with the values, in MCT1 and MCT2, $n_{77 \text{ K}} = 6 \times 10^{16} \text{ cm}^{-3}$ and $4 \times 10^{16} \text{ cm}^{-3}$, respectively, one order of magnitude above those before irradiation (Table I).

III. LIFETIME RESULTS BEFORE AND AFTER ELECTRON IRRADIATION

Before irradiation (Fig. 1), the average lifetime τ is 278 ± 1 ps at 77 K and 280 ± 1 ps at 300 K. After 20 K irradiation at the fluence of $10^{18} e^- \text{cm}^{-2}$ and annealing up to 77 K, the average positron lifetime at 77 K in MCT2 increases by 25 ps from 278 ± 1 to 303 ± 1 ps (Fig. 2). The main annealing stage occurs from 200 to 280 K where the lifetime increases up to 312 ± 1 ps. The spectra have two components. The two lifetime components can, however, be resolved with a good accuracy only for annealings above 230 K. The long component τ_2 is 323 ± 5 ps. Its intensity increases between 200 and 280 K during the annealing stage (Fig. 2) from 75% to 88%.

The average positron lifetime τ in MCT2 was measured as a function of temperature after annealing at 205, 255,

and 340 K, respectively. Figure 3 shows that the lifetime remains nearly constant between 80 and 180 K after 205 K as well as after 255 and 340 K annealings. Above 180 K and after annealing at 340 K, strong reversible effects are observed in Fig. 3: the average positron lifetime displays a strong decrease when the temperature increases from 260 to 330 K.

The long lifetimes as a function of measurement temperature in Fig. 3 show a constant value of 323 ± 5 ps after the 205 and 255 K annealings. After annealing at 340 K, the decomposition of the spectra appears temperature dependent. The long lifetime τ_2 is first constant with a value of 327 ± 5 ps up to 240 K and then decreases reversibly from 327 ± 5 to 306 ± 5 ps between 240 and 300 K.

Aging of MCT2 at room temperature for several weeks induces a decrease in the average lifetime τ . τ decreases from 312 ± 1 to 295 ± 1 ps at 77 K and from 295 ± 1 to 285 ± 1 ps at 300 K. After aging, the long lifetime τ_2 at 77 K decreases from 323 to 309 ps (full circles in Fig. 3). It is independent of measurement temperature (Fig. 3) and remains equal to 309 ± 5 ps from 20 to 160 K. Above 160 K, the spectra are resolved in only one component.

After irradiation of MCT1 at the fluence 5×10^{18}

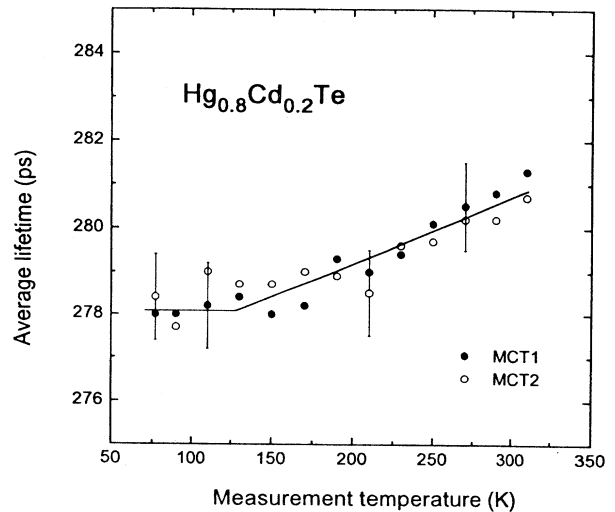


FIG. 1. The temperature dependence of the average positron lifetimes τ ($\Delta\tau = \pm 1$ ps) before electron irradiation in *n*-type $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ with $n_{77 \text{ K}} = 3 \times 10^{15} \text{ cm}^{-3}$.

$e^- \text{cm}^{-2}$, the average lifetime at 77 K increases up to 301 ± 1 ps. The main annealing stage starts at about 200 K where the average lifetime increases. This annealing stage is similar to that observed in MCT2 for the fluence $10^{18} e^- \text{cm}^{-2}$. Figure 4 shows that after annealing at 340 K the temperature dependences of the average lifetime τ and long lifetime τ_2 in MCT1 are similar to those observed in MCT2. The decrease of the long lifetime τ_2 from 325 ± 5 to 310 ± 5 ps is, however, shifted at lower temperature by about 40 K.

IV. IRRADIATION-INDUCED VACANCIES

The average positron lifetime is the sum of the annihilation lifetimes in the different positron states weighted by the respective fractions of positrons annihilating in these states. Positrons are trapped at neutral and negative vacancy-type defects. The positron lifetime is then

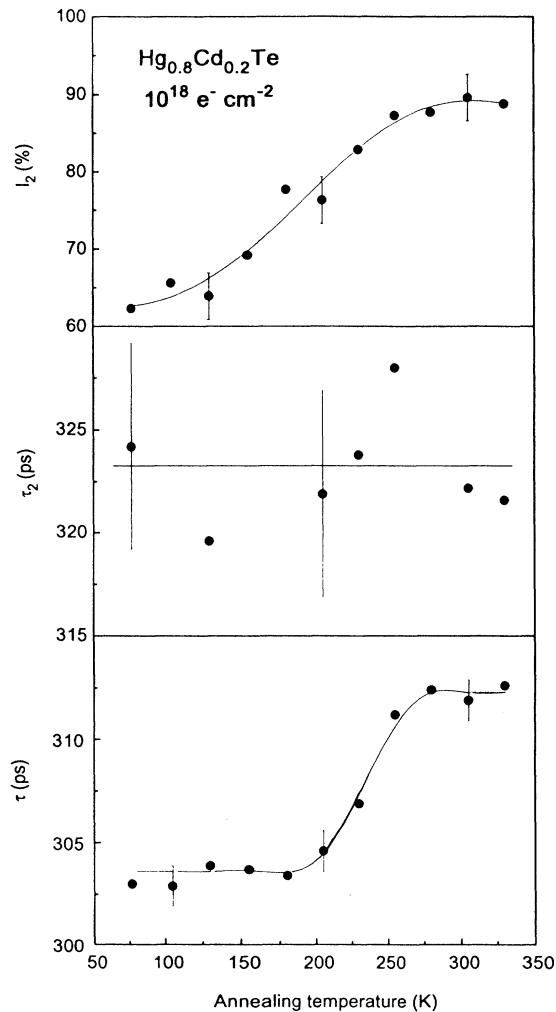


FIG. 2. Average positron lifetime τ ($\Delta\tau = \pm 1$ ps), trapped positron lifetime τ_2 , and its intensity I_2 ($\Delta I_2 = \pm 5\%$) as a function of annealing temperature in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ irradiated with 2.5 MeV electrons at a fluence of $10^{18} e^- \text{cm}^{-2}$.

longer^{17–23} than when they are delocalized in the lattice due to the lower electron density at vacancy-type defects. Positron trapping at vacancies thus manifests itself in an increase of the positron lifetime.

Before irradiation, the lifetime values (Fig. 1) and their temperature dependence are characteristic of those we find in stoichiometric annealed samples and are attributed to positron annihilation in the lattice.²⁶ The small increase of the positron lifetime with temperature is partly an effect of the lattice thermal expansion. The lifetime of 280 ps at 300 K falls in the range of values which have been earlier proposed for annihilation in the lattice.^{15,16}

After annealing at 77 K, the average lifetime at 77 K is definitely longer than before irradiation. The increase of the average lifetime by 25 ps indicates positron trapping at vacancy-type defects after irradiation. The long lifetime τ_2 323 ± 5 ps is observed in both investigated samples independent of their different irradiation doses (Fig. 4) and over a wide range of annealing and measurement temperatures. This leads us to conclude that the long lifetime τ_2 323 ps characterizes the main vacancy-type defect trapping positrons after irradiation. This lifetime

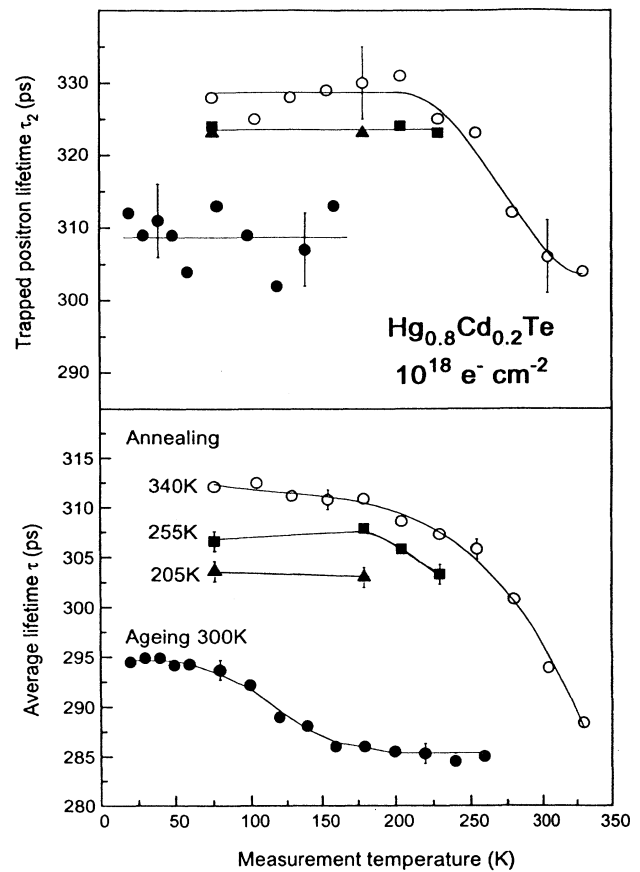


FIG. 3. Temperature dependence of the average positron lifetime and the trapped positron lifetimes after annealing stages at 205 K (\blacktriangle), 255 K (\blacksquare), and 340 K (\circ) and aging (\bullet) at room temperature over several weeks in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ irradiated with 2.5 MeV electrons at a fluence of $10^{18} e^- \text{cm}^{-2}$.

TABLE II. Theoretical and experimental values for positron lifetimes at 77 K in lattice τ_b , at vacancies and first nearest neighbor (1NN) divacancies assuming no lattice distortion (Ref. 27) and for $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ linear dependence of the positron lifetimes.

	HgTe		CdTe		$\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$			
	Calculated		Calculated		Calculated		Experimental	
τ_b (ps)	274		292		278		278±1	
Monovacancy	V_{Hg}	V_{Te}	V_{Cd}	V_{Te}	$V_{\text{Hg,Cd}}$	V_{Te}	$V_{\text{Hg,Cd}}$	V_{Te}
τ (ps)	304	315	321	339	307	320	309±5	323±5
1NN divacancy	$V_{\text{Hg}}-V_{\text{Te}}$		$V_{\text{Cd}}-V_{\text{Te}}$		$V_{\text{Hg,Cd}}-V_{\text{Te}}$			
τ (ps)	362		384		366			

is about 17% longer than the lifetime in the lattice. Such an increase when compared to the values calculated in CdTe and HgTe (Ref. 27) for trapping at V_{Te} and V_{Cd} or V_{Hg} monovacancies and $V_{\text{Cd}}-V_{\text{Te}}$ or $V_{\text{Hg}}-V_{\text{Te}}$ divacancies corresponds to the open volume of a monovacancy (Table II).

In addition to the irradiation-induced lifetime of 323 ps, we observe that positron trapping after aging (Fig. 3) gives rise to the long lifetime τ_2 of 309±5 ps. During this aging of several weeks at room temperature, the 323 ps lifetime disappears irreversibly and only the lifetime of 309±5 ps is detected. Lifetimes of about 305–310 ps have earlier been observed in as-grown crystals and attributed to V_{Hg} vacancies.^{15,16} We then conclude that irradiation has introduced V_{Hg} vacancies. Table II shows that there is good agreement between the experimental and theoretical values of the lifetimes at V_{Hg} .

In Fig. 4 we notice that at low temperatures the decomposition of the spectra is nearly the same although the irradiation fluence differs by a factor of 5. This may indicate that for these fluences we are already in a region where the production of defects is near its saturation level.

V. CHARGE STATES OF THE VACANCIES

We examine below successively the charge state of the 309 and 323 ps vacancies.

After aging at 300 K, we can check from the decomposition of the spectra in Fig. 4 whether only V_{Hg} vacancies trap positrons. It is sufficient to compare the lattice lifetime τ_b to the lifetime τ_s calculated with

$$\tau_s(T) = \left[\frac{I_1}{\tau_1} + \frac{I_2}{\tau_2} \right]^{-1} \quad (1)$$

In the framework of the positron trapping model these two quantities are equal when positrons annihilate with only one other lifetime than that in the lattice.²⁸ From 20 to 160 K, Fig. 5 shows that, after aging, one can consider that τ_s and τ_b are nearly equal. We can then conclude that mainly V_{Hg} are trapping positrons and, consequently, calculate the positron rate at V_{Hg} , $k_V(T)$, by the relation

$$k_V(T) = \lambda_b(T) \frac{\tau(T) - \tau_b(T)}{\tau_2 - \tau(T)} \quad (2)$$

where $\lambda_b(T)$ is the annihilation rate in the lattice, $\lambda_b(T) = \tau_b^{-1}(T)$. The trapping rate at V_{Hg} , $k_V(T)$, decreases reversibly as the temperature increases as shown in Fig. 6. Such a decrease is unlikely to be due to change of the charge state of V_{Hg} . The electron concentration is so high that the Fermi level remains well above the ionization levels of V_{Hg} , which are believed to be near the

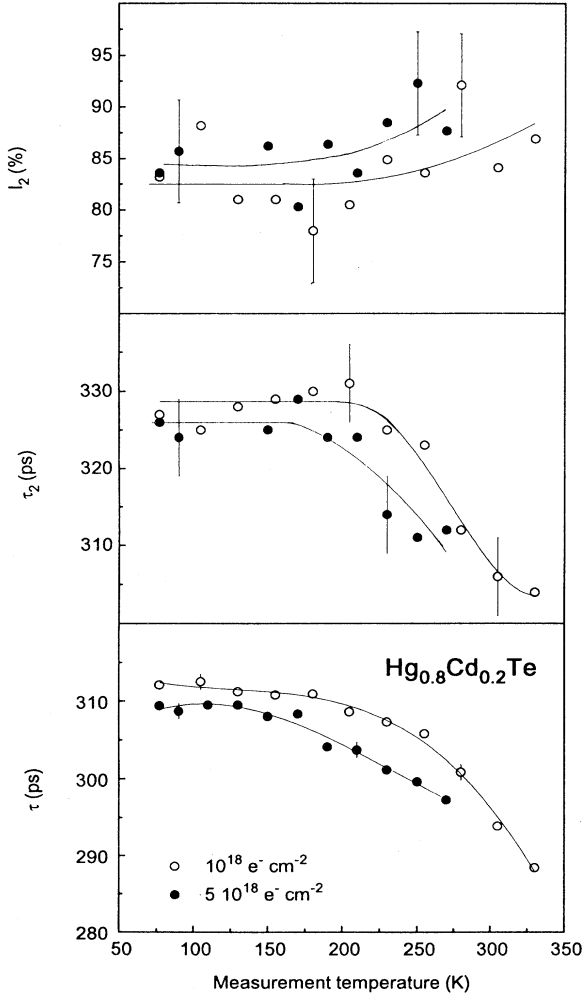


FIG. 4. Average positron lifetime τ ($\Delta\tau = \pm 1$ ps), trapped positron lifetime τ_2 , and its intensity I_2 ($\Delta I_2 = \pm 5\%$) as a function of measurement temperature after annealing to 340 K in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ irradiated with 2.5 MeV electrons at a fluence of 10^{18} and $5 \times 10^{18} e^- \text{cm}^{-2}$.

valence-band edge.²⁹ The charge state of V_{Hg} remaining constant as the temperature increases from 20 to 300 K, the decrease of $k_V(T)$ rather reflects a decrease of the positron trapping coefficient at V_{Hg} . Such a decrease is the fingerprint of negatively charged vacancies²⁷ and allows us to conclude that V_{Hg} vacancies are negative, in agreement with their acceptor properties.^{4,15,16}

After annealing at 340 K, we observed that as a function of temperature the vacancy lifetime τ_2 decreases reversibly from about 327 to 306 ± 5 ps (Fig. 4). The lifetime value of 306 ± 5 ps is of the same order as those

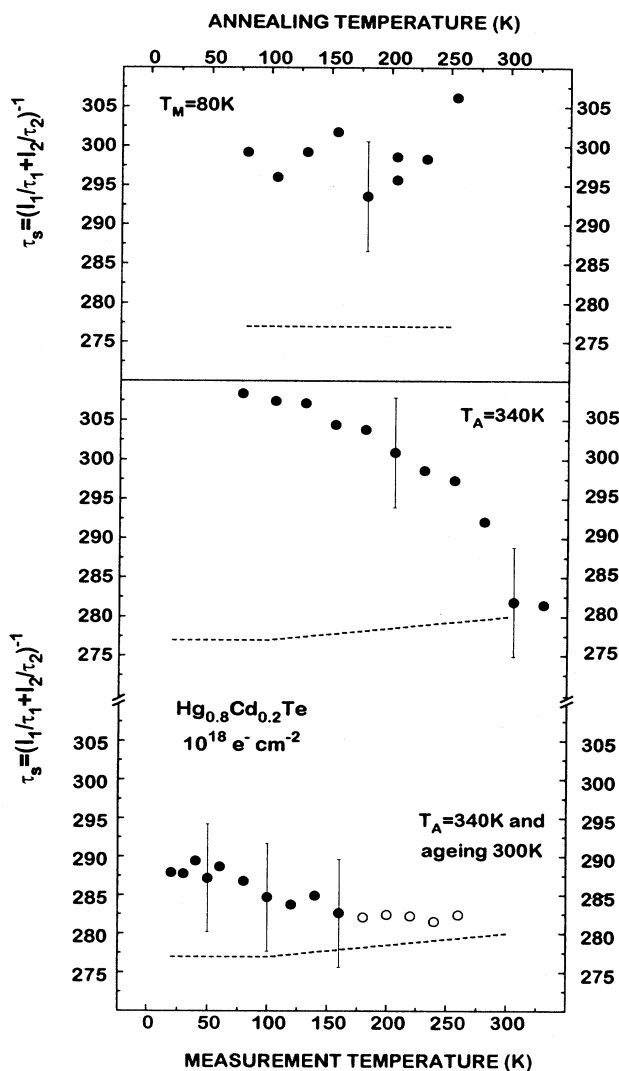


FIG. 5. Calculated lifetime from the decomposition of the lifetime spectra $\tau_s = (I_1/\tau_1 + I_2/\tau_2)^{-1}$ as a function of the annealing temperature, as a function of the measurement temperature just after annealing at 340 K and after annealing at 340 K and subsequent aging at 300 K for $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ irradiated with 2.5 MeV electrons at a fluence of $10^{18} e^- \text{cm}^{-2}$. The open circles correspond to the decomposition where the long lifetime is fixed to the mean value 305 ps. The dotted lines correspond to the lifetime in lattice.

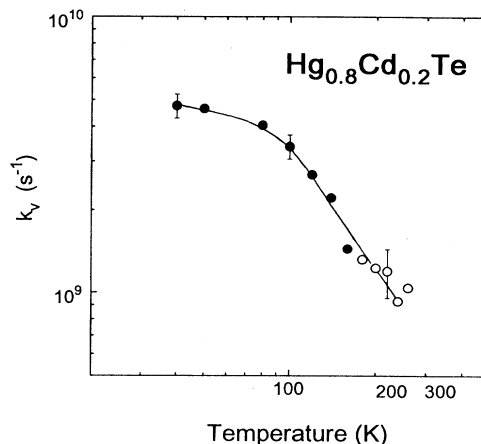


FIG. 6. Positron trapping rate as a function of temperature after annealing at 340 K and aging at 300 K for $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ irradiated with 2.5 MeV electrons at a fluence of $10^{18} e^- \text{cm}^{-2}$. The open circles correspond to the decomposition where the long lifetime is fixed to the mean value 305 ps.

found after aging and attributed to V_{Hg} . This suggests that positron trapping takes place in V_{Hg} at the end of this reversible transition. The question is whether the 323 ± 5 ps lifetime arises also from positron trapping at V_{Hg} . If so, we expect that the 323 ps lifetime corresponds to a more negatively charged state of V_{Hg} than the lifetime 309 ps because the Fermi level is higher than after aging. There is evidence for a neutral charge state of the 323 ps vacancy rather than for a negative one. This comes from the flat temperature dependence of the intensity of the lifetime 323 ps in Fig. 4 which leads us to conclude that the positron trapping coefficient at the 323 ps vacancy is likely constant from 77 to 200 K. This property characterizes trapping at neutral vacancies according to theory²⁷ or experiment.¹⁷ It seems then unlikely that the 323 ps vacancy is related to V_{Hg} . The other possibility is that it is related to V_{Te} . In Table II, the calculated lifetime at V_{Te} is indeed longer than at V_{Hg} and much closer to the value 323 ps than the lifetime calculated at the divacancy.

After annealing at 340 K, the calculated lifetime τ_s [see Eq. (1)] from the decomposition of the spectra as a function of temperature in Fig. 4 becomes equal to τ_b when the 309 ps lifetime is detected. It occurs above 240 K after $5 \times 10^{18} e^- \text{cm}^{-2}$ irradiation and, as seen in Fig. 5, above 280 K after $10^{18} e^- \text{cm}^{-2}$ irradiation. This means that then only V_{Hg} vacancies trap positrons. We propose that the reversible transition 323 to 309 ps as a function of temperature in Fig. 4 is a Fermi-level-controlled one. The trapping at the V_{Te} vacancy is lost when the Fermi level goes down in the gap because the V_{Te} vacancy becomes positively charged. Only trapping at V_{Hg} is then left. The electron concentrations are higher than $4 \times 10^{16} \text{ cm}^{-3}$ when V_{Te} vacancies are observed, which suggests that the V_{Te} level 0/+ is high in the conduction band. At low temperature when the Fermi level is pinned by the

ionization level of the donor defect according to Ref. 13, most likely the V_{Te} vacancy we observe here, there is competition between the trappings at V_{Te} and V_{Hg} . There is, however, no evidence that the 323 ps is a mixing of two different lifetimes. On the contrary, as mentioned above, it appears to be due only one type of defect.

It is a surprising conclusion at first glance that neutral vacancies can compete with negatively charge vacancies, especially at low temperature. The trapping coefficient at negative vacancies is indeed expected to be higher by an order of magnitude at 20 K than that at neutral vacancies according to calculations.²⁷ This necessitates concentrations of V_{Te} vacancies higher than V_{Hg} vacancies. We are led to conclude that the introduction of vacancy donor levels appears, therefore, much higher than that of vacancy acceptor levels. Such a dissymmetry may partly explain why one observes that irradiation in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ materials produces high electron concentration.¹⁴

During aging at 300 K, V_{Hg} recovery takes place. This recovery is reflected in Fig. 3 by the decrease of the average lifetime at 300 K from 295 to 285 ps, due to the decrease of the fraction of the positrons trapped at the 309 ± 5 ps. It is unclear whether the loss of the 323 ps lifetime after aging means that during aging the V_{Te} vacancy disappears also. During aging, the Fermi level goes down. The positron signal due to the V_{Te} vacancy may simply disappear because the vacancy become positive.

VI. NEGATIVE IONS

The τ_s values calculated from expression (1) for the decompositions with the lifetime 323 ps are longer than the τ_b values. This is illustrated in Fig. 5 for MCT2. It follows that at least one other type of defect traps positrons in addition to the 323 ps vacancy. This indicates that, at low temperature, other types of defects are trapping the positrons in addition to the defect with the 323 ps characteristic lifetime.

The increase of the intensity of the 323 ps lifetime in Fig. 2 between 150 and 280 K indicates that the fraction of positrons trapped at the 323 ps vacancy progressively increases as a function of annealing temperature. A simple interpretation is to propose that the concentration of 323 ps vacancies increases as a function of annealing tem-

perature. However, we find it difficult to explain how annealing can produce higher concentration of 323 ps monovacancies. On the other hand, it seems unlikely that the V_{Hg} vacancies disappear since they are still observed after annealing at 340 K or aging at 300 K. Consequently, we prefer the following interpretation for which the concentration of 323 and 309 ps vacancies can remain constant or even decrease. We propose that, in addition to the 323 and 309 ps vacancies, irradiation introduces negative ions. The trapping at vacancy-type defects and negative ions is competing at 77 K. Positrons trapped at negative ions annihilate with a lifetime close to the lifetime in the lattice and so shorter than at vacancy-type defects. Between 150 and 280 K we can attribute the increase of the average lifetime to the recovery of the negative ions. The negative ions disappearing, more positrons become trapped at vacancies which causes the increase of the average lifetime. Among the defects in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ which may be negatively charged ions in *n*-type materials are Te interstitials, according to calculations.

In previous studies, recovery stages for the resistivity and the carrier concentration¹² have also been observed in the range 150–200 K. The electron concentration was decreasing in this recovery which means that, if our interpretation is correct, the negative ions recombine with donor defects. As Te interstitials are expected to recombine with V_{Te} vacancies, our identification of the negative ions as Te interstitials is consistent with the loss of the 323 ps lifetime during aging at 300 K and suggests that in addition to a Fermi-level effect (see above) this loss is due to the disappearance of V_{Te} vacancies.

VII. CONCLUSION

Positron lifetime measurements give evidence that electron irradiation introduces vacancies in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ which give rise to two lifetimes, 323 ± 5 and 309 ± 5 ps. The 309 ps vacancies survive at room temperature and are slowly eliminated by aging over several weeks. The positron-trapping coefficient decreases with increasing temperature, indicating that the 309 ps vacancies are negatively charged. The 309 ps vacancies which have been observed earlier are identified with the V_{Hg} vacancies. The 323 ps lifetime is observed when the Fermi level is high in the gap and is tentatively attributed to V_{Te} vacancies.

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