Study of *DX* center in $Cd_{0.8}Zn_{0.2}Te$:Cl by positron annihilation

S. Fung,[a] Y. Y. Shan, A. H. Deng, C. C. Ling, and C. D. Beling
Department of Physics, The University of Hong Kong, Hong Kong

K. G. Lynn
Department of Physics, Washington State University, Pullman, Washington 99163

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Variable energy positron beam and positron annihilation lifetime experiments have been carried out to study the *DX* center in $Cd_{0.8}Zn_{0.2}Te$:Cl at 50 K. A short positron effective diffusion length of 275 ± 25 Å and a large intensity of $79.0\%\pm0.3\%$ for the long lifetime component indicate a strong trapping effect at *DX* centers. A trapping rate of $\kappa=1.53\pm0.05\times10^{9}$ s^{-1} and a positron lifetime of 335 ± 2 ps at the *DX* center were obtained. The concentration of *DX* centers is found to be $5.9\pm0.7\times10^{16}$ cm^{-3}, which is in good agreement with the results obtained using Hall effect and thermo-electric effect measurements. © *1998 American Institute of Physics.* [S0021-8979(98)02216-6]

I. INTRODUCTION

Defects having deep levels in the band gap play critical roles in the electrical and optical properties of semiconductors. One group of deep defects that has been extensively studied for many years is the *DX* centers.[1,2] Upon photoexcitation at low temperatures, semiconductor compounds having *DX* centers exhibit persistent photoconductivity (PPC). Because of this effect, *DX* centers can be used as an optically controllable source of electrons,[3–5] and hence have great potential applications in optical writing and high-density data storage.[4–7] In most III-V compounds having *DX* centers such as AlGaAs, written memory persists only below $T_A\sim50-100$ K, the annealing temperature of the PPC. However, in II-VI semiconductor compounds, where *DX* centers show properties similar to those in AlGaAs,[8,9] much higher annealing temperatures of *DX* centers can be obtained.[10,11] Although some optical, electrical, and microscopic structure properties of *DX* centers have been studied extensively, not many direct observations giving information about the concentration, charge states, and microstructures of *DX* centers in $Cd_{0.8}Zn_{0.2}Te$:Cl have been reported.

In this work, positron annihilation techniques have been employed to study the *DX* center in $Cd_{0.8}Zn_{0.2}Te$:Cl. The positron is one of the few nondestructive sensitive probes for defect studies in condensed matter.[12,13] This results from their propensity to be trapped at neutral and negative vacancy defect sites in the lattice. Since the electron density at these defects is lower than that in the bulk, the lifetime of positrons is longer than that in the perfect crystal. Moreover, the low electron momentum distribution experienced by positrons is enhanced at the vacancy sites, reducing the Doppler broadening of the 511 keV annihilation line. Consequently, positron lifetime and Doppler-broadening measurements give defect information on an atomic scale.

Recently positron trapping and the microscopic structure of *DX* centers in $Cd_{0.8}Zn_{0.2}Te$:Cl with deep donor Cl atoms relaxing towards their interstitial position along the [111] direction have been reported.[14,15] However, few results of the positron lifetime related to this interstitial relaxation have been obtained. In this work, positron diffusion and trapping in $Cd_{0.8}Zn_{0.2}Te$:Cl at low temperature have been studied by means of a variable energy positron beam and positron lifetime spectroscopy. We also show that, in comparison to the Hall effect or thermo-electrical effect measurements, the concentration of *DX* centers can be more directly determined, where free carrier concentrations were measured to deduce the *DX* center concentration after they had been excited to d^+ states.[10,11,16]

II. EXPERIMENT

The $Cd_{0.8}Zn_{0.2}Te$ single crystal used in this study was grown by the vertical Bridgman technique.[17] Chlorine doping was achieved by adding ZnCl to the melt. The sample was annealed in Cd vapor at 0.02 atm at 600 °C for 5 days to remove Cd(Zn) vacancies and *A* centers, i.e., Cd(Zn) vacancy–Cl atom pairs (V_{Cd}–Cl_{Te}).

Positron diffusion and trapping experiments were carried out using a monoenergetic positron beam and a standard fast–fast lifetime spectrometer.[13,18] Both positron beam and positron lifetime measurements were carried out in the dark at 50 K to keep the *DX* centers in their deep stable state, which has a configuration that the Cl donor atoms relax to the interstitial positions along the [111] direction from their substitutional positions.[14] The sample was mounted on a copper cold finger of a closed-cycle He refrigerator cryostat and the pressure during the measurements was 10^{-7} mbar. For the positron beam measurements, a high-purity Ge gamma detector was used to detect the Doppler broadened positron–electron annihilation gamma spectra. A total of 1×10^{6} counts was collected under the 511 keV annihilation photopeak using a digitally stabilized multichannel analyzer. For the positron lifetime measurements, 2–3 μCi of ^{22}NaCl positron source was directly deposited onto the sample to reduce any errors associated with source annihilation correc-

[a]Electronic mail: sfung@hkucc.hku.hk

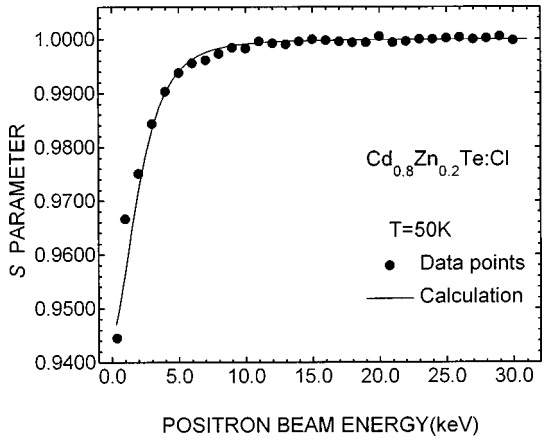

FIG. 1. Line shape S parameter as a function of the incident positron energy at 50 K under dark conditions. The S values were normalized to the bulk S value. The solid line is the calculation of the diffusion–annihilation model for effective diffusion length $L_{\text{eff}}=275$ Å.

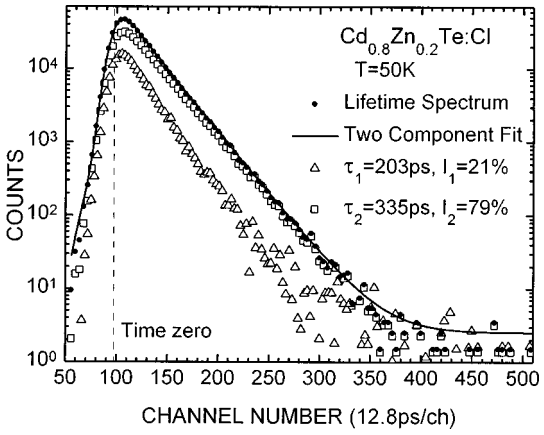

FIG. 2. The positron–annihilation lifetime spectra of $Cd_{0.8}Zn_{0.2}Te$:Cl in the dark at 50 K.

tion. The time resolution of the lifetime spectrometer was 235 ps full width half maximum (FWHM), and each spectrum contained 2×10^6 events.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the measured positron annihilation S parameter as a function of the incident positron beam energy in the dark at 50 K. At the very low incident energy of 0.1 keV, a S value of 0.9440 characterizing positrons annihilating mainly on the surface was observed. As the energy increases, positrons are implanted deep into the sample. The S parameter reaches a saturation value when the beam energy increases to above 10 keV, indicating that positrons annihilate in the bulk region. All the S values in Fig. 1 are normalized to this saturating bulk S value. Since positron diffusion in the sample is limited by positron trapping into the defect trapping centers, the measurements of effective positron diffusion length and diffusion coefficient give the properties of the trapping centers. By solving the positron diffusion–annihilation equation, the fraction of positrons diffusing to the surface is determined to be:[19,20]

$$f_s(E)=\int_0^{+\infty} P_E(z)e^{-z/L_{eff}}dz, \tag{1}$$

where

$$P_E(z)=m\frac{z^{m-1}}{z_0^m}\exp\left[-\left(\frac{z}{z_0}\right)^m\right], \tag{2}$$

is the positron beam implantation profile, $z_0=(\alpha/\rho)E^n$, where the beam energy E is in keV and ρ ($=5.823$ g cm^{-3} for $Cd_{0.8}Zn_{0.2}Te$) is the density of the sample. The parameters are taken as $\alpha=450$ Å g cm^{-3} keV^{-n}, where $m=2.0$, and $n=1.6$.[21–23] The positron mean implantation depth is $z_0\Gamma(1+1/m)$. The effective diffusion length,

$$L_{\text{eff}}=\sqrt{D_+/\lambda_{\text{eff}}}, \tag{3}$$

depends on the positron diffusion coefficient D_+, and the effective positron annihilation rate $\lambda_{\text{eff}}=\lambda_b+\kappa$. λ_b is the annihilation rate of the bulk, and κ is the defect trapping rate. The measured line shape parameter $S(E)$ can be modeled as:[24]

$$S(E)=f_s(E)S_s+[1-f_s(E)]S_b. \tag{4}$$

By fitting (4) to the experimental data, the effective diffusion length $L_{\text{eff}}=275\pm25$ Å is obtained. This extremely short diffusion length indicates that positron diffusion is limited by trapping effects in the bulk. It has been observed that positrons are trapped and annihilate at DX centers before photoexcitation and at chlorine A centers after photoexcitation at low temperatures.[15] The implanted positrons were considered to be trapped mainly at the DX centers here in our measurements carried out in the dark at 50 K. Therefore the concentration of DX centers can be directly determined by measuring its positron trapping rate. The trapping rate, κ, can be obtained from the positron annihilation lifetime measurements and using the two-state trapping model.[25]

In the positron lifetime study, the lifetime spectra were analyzed using the data analysis package POSFIT-88.[26] To obtain the time resolution function, the RESOLUTION code[26] was used to deconvolute the natural 182 ps lifetime spectrum of ^{207}Bi.[27] It was found that a linear combination of two Gaussians gave a good fit to the resolution function of the system. One Gaussian has a FWHM value of 235 ps and intensity of 95%. The other has a FWHM value of 472 ps, intensity of 5%, and 35 ps left shift from the time zero. In a defect free crystal, positrons are delocalized and annihilate with a single lifetime component. When there are defect trapping centers, the lifetime spectrum can be fitted by the sum of exponentials. In our case, the two-state trapping model describes the system.[25] The lifetime spectrum can be fitted well by the sum of two exponential components convoluted with the system resolution function $G(t)$:

$$S(t)=N_0\int_{-\infty}^{+\infty}[I_1\lambda_1e^{-\lambda_1 v}+I_2\lambda_2e^{-\lambda_2 v}]G(t-v)dv. \tag{5}$$

Figure 2 shows the measured positron annihilation lifetime spectrum and its decomposition. $\tau_1(1/\lambda_1)$ and $\tau_2(1/\lambda_2)$ are found to be 203 ± 2 and 335 ± 2 ps, respectively, from free fitting. The intensities of the two components, I_1 and I_2,

are 21.0%±0.3% and 79.0%±0.3%, respectively. Using the two-state trapping model, the delocalized positron lifetime in $Cd_{0.8}Zn_{0.2}Te$ can be determined as $\tau_b=(I_1/\tau_1+I_2/\tau_2)^{-1}=294$ ps, which is in agreement with the theoretical positron lifetime value of 290 ps in CdTe.[28] Since few experimental results of positron lifetime in $Cd_{1-x}Zn_xTe$ have been reported, we believe that the somewhat larger value of τ_b compared to that in CdTe comes from the Zn composition replacing the Cd composition. The intensity I_2 of 79% for the observed long component indicates that a large number of positrons are trapped at defects. The positron lifetime at defects is 335 ps, slightly longer than the free state lifetime in bulk. This can be explained by the *DX* center configuration where the Cl donor atoms relax to the interstitial positions along the [111] direction creating a vacancylike volume and hence a longer lifetime value is observed when positrons are trapped at the *DX* centers.

The positron trapping rate from the delocalized state into the *DX* center can be obtained as:

$$\kappa=\lambda_1-\lambda_b=I_2(\lambda_1-\lambda_2). \tag{6}$$

Taking I_2 to be 79%, λ_1 to be $(203\ \text{ps})^{-1}$, and λ_2 to be $(335\ \text{ps})^{-1}$, the trapping rate κ of $1.53\pm0.05\times10^9\ \text{s}^{-1}$ is obtained. From (3), the positron diffusion coefficient, $D_+=L_{\text{eff}}^2(\lambda_b+\kappa)=3.7\pm0.6\times10^{-2}\ \text{cm}^2\ \text{s}^{-1}$, is obtained. The trapping rate is proportional to the defect concentration *C*:

$$\kappa=\mu_d C, \tag{7}$$

where μ_d is the position trapping coefficient. It has been reported that the chlorine *DX* center has the microscopic structure of a Cd–Cl bond with a separation of 5.5 Å compared to the ''normal'' Cd–Cl bond length of 2.8 Å for the tetrahedral substitutional d^+ state.[14] This increase of bond separation comes from the relaxation of the donor Cl atom to the interstitial position from its substitutional position and might be seen as a vacancylike positron trapping center. The trapping coefficient is given by $\mu_d=4\pi RD_+$.[29] Taking the vacancy size *R* to be 5.5 Å, and the diffusion coefficient D_+ to be $3.7\pm0.6\times10^{-2}\ \text{cm}^2\ \text{s}^{-1}$, the trapping coefficient μ_d is deduced to be $2.6\pm0.4\times10^{-8}\ \text{cm}^3\ \text{s}^{-1}$. Consequently, the *DX* center concentration of $5.9\pm0.7\times10^{16}\ \text{cm}^{-3}$ is obtained. The concentration of the *DX* center can also be estimated from (7) by taking the trapping coefficient of a vacancy, μ_d, to be approximately $1\times10^{15}\ \text{s}^{-1}/N_{\text{at}}$, where N_{at} is the atomic density.[30,31] Taking the atomic density to be $3.05\times10^{22}\ \text{cm}^{-3}$ for $Cd_{0.8}Zn_{0.2}Te$, the trapping coefficient and the *DX* center concentration are determined to be $3.3\times10^{-8}\ \text{cm}^3\ \text{s}^{-1}$ and of $4.6\pm0.2\times10^{16}\ \text{cm}^{-3}$, respectively, which have the same order of magnitude as those obtained from the diffusion data. Since the chlorine *DX* center only has a vacancylike configuration, the deviation between its exact positron trapping coefficient and that of the vacancy used above could possibly cause some uncertainties in the determination of the *DX* center concentration. The results we obtained using positron annihilation techniques in this study are in good agreement with those from Hall effect measurements and thermoelectric effect current measurements.[10,11,16] In these measurements, the *DX* center concentration of $\sim6.0\times10^{16}\ \text{cm}^{-3}$ was deduced indirectly by measuring the free electron concentration in the conduction band when the *DX* centers were photoexcited to their metastable d^+ states.

IV. CONCLUSIONS

In summary, positron diffusion and positron trapping into chlorine *DX* centers in $Cd_{0.8}Zn_{0.2}Te$ have been studied by means of low energy positron beam and positron lifetime spectroscopy at 50 K. The extremely short diffusion length of 275 Å and the large intensity of the defect lifetime component of 79% indicate strong positron trapping at the *DX* centers. The observed positron lifetime of 335 ps at the chlorine *DX* center, in which the Cl atom has relaxed to the interstitial position to form a vacancylike trapping center, is longer than the delocalized positron lifetime of 294 ps. The positron trapping rate and the concentration of *DX* center are found to be $1.53\pm0.05\times10^9\ \text{s}^{-1}$ and $5.9\pm0.7\times10^{16}\ \text{cm}^{-3}$, respectively. The present study shows that, in comparison with more conventional methods, the concentration of defects can be more directly determined by the use of positron annihilation techniques.

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