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Positron lifetime measurements on neutron-irradiated InP crystals

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Neutron-irradiated InP single crystals have been investigated by positron-lifetime measurements. The samples were irradiated with thermal neutrons at different fluences yielding concentrations for Sn-transmuted atoms between 2×10^{15} and 2×10^{18} cm^{-3}. The lifetime spectra have been analyzed into one exponential decay component. The mean lifetimes show a monotonous increase with the irradiation dose from 246 to 282 ps. The increase in the lifetime has been associated to a defect containing an Indium vacancy. Thermal annealing at 550 °C reduces the lifetime until values closed to those obtained for the as-grown and conventionally doped InP crystals. © *1996 American Institute of Physics.* [S0021-8979(96)03711-5]

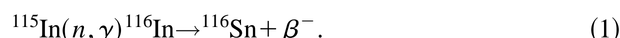
I. INTRODUCTION

Positron annihilation spectroscopy (PAS) has become a valuable tool to study the properties in solids.[1] In particular, positron techniques have demonstrated its capability on analyze the defect structure on semiconductor materials.[2,3] At present, the positron annihilation is successfully applied to investigate the structure of the native defects in compound III–V semiconductors.[4–6] However, few works have been reported in InP, which have attracted much attention because it is one of the most promising materials for the development of optoelectronics.

In this material, the positron lifetime technique has been applied to investigate the nature of the native defects in as-grown InP.[7,8] It is found that the experimental bulk positron lifetime values published vary from 235 to 247 ps. Recently, Bretagnon *et al.*[8] have reported a systematic study of grown-in defects in crystals doped with different dopants and carrier concentrations. These authors have resolved a second lifetime, between 263 and 272 ps which has been attributed to positrons annihilating at indium vacancy-impurity related defects.

Neutron-transmutation doping (NTD) is a convenient method for semiconductor doping purposes when a precise amount and an homogeneous distribution of the impurities are required. NTD relies on the capture of thermal neutrons by the host lattice nuclei. The process depends on the capture cross-sections and the natural abundance of each isotope. It has been shown that InP can be effectively doped by neutron transmutation, yielding a precise concentration of doping atoms which act as donors and control the electron concentration.[9,10]

The main impurity introduced in InP by NTD is Sn, which comes from the transmutation of the most naturally abundant ^{115}In isotope (95.7%) according the following reaction:



$$^{115}\mathrm{In}(n,\gamma)^{116}\mathrm{In}\rightarrow{}^{116}\mathrm{Sn}+\beta^{-}. \quad (1)$$

^{115}In has a high neutron capture cross section ($\sigma_c=199$ b). The rest of the isotopes of InP have the following neutron capture cross-sections and natural abundance: 10.7 b and 4.3% for ^{113}In and 0.19 b and 100% of abundance for ^{31}P. ^{113}In transmute mainly in Sn and ^{31}P into S. Among all the nuclear reactions, 99.9% transmute In to Sn and about 0.1% transmute P into S atoms. The values of neutron capture cross-sections and natural abundances were extracted from.[11]

During the NTD process some lattice defects are created due to the γ and β rays produced in the nuclear reaccions and the fast neutrons which unavoidable accompany the thermal neutron flux and cannot be completely eliminated. Therefore, a suitable annealing step has to be performed in order to recover the crystal lattice. In the case of InP, annealing at 550 °C is enough to electrically activate the doping impurities. This has been achieved using both classical (10 min) and rapid (10 s) thermal annealing methods.[9]

We present here lifetime positron measurements in unirradiated and neutron-irradiated InP single crystals and the evolution of their lifetime parameters as a function of the calculated Sn concentration. The main purpose of this work is to study the nature of radiation induced defects affecting the positron annihilation lifetime and their recover after thermal annealing.

II. EXPERIMENT

The starting material used were undoped n-type LEC-grown InP, with a free carrier density of about $(2–4)\times10^{15}$ cm^{-3} at room temperature. Samples of approximated dimensions 7×7 mm^2 and 350 μm thick were extracted from the same wafer and subsequently irradiated within a low flux nuclear reactor with several doses of thermal neutrons in order to obtain additional doping concentrations ranging

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TABLE I. Irradiation conditions and expected Sn concentrations in the samples used in this work. Samples 10 and 11 were annealed at 550 °C with a residence time of 10 min and then measured at room temperature.

Ref.	Sample	Irradiation parameters		Observations
		Flux $[cm^{-1} s^{-1}]$	Fluence $[cm^{-2}]$	
(1)	as-grown			
(2)	NTD:[Sn]=2×10^{15}	10^{12}	5.30×10^{14}	
(3)	NTD:[Sn]=4×10^{15}	10^{12}	1.06×10^{15}	
(4)	NTD:[Sn]=1×10^{16}	10^{12}	2.65×10^{15}	
(5)	NTD:[Sn]=3×10^{16}	10^{12}	7.95×10^{15}	
(6)	NTD:[Sn]=1×10^{17}	10^{12}	2.65×10^{16}	
(7)	NTD:[Sn]=3×10^{17}	4.4×10^{11}	7.95×10^{16}	
(8)	NTD:[Sn]=1×10^{18}	4.7×10^{11}	2.65×10^{17}	
(9)	NTD:[Sn]=2×10^{18}	4.4×10^{11}	5.30×10^{17}	
(10)	NTD:[Sn]=10^{17}	10^{12}	2.65×10^{16}	550 °C, 10 min
(11)	NTD:[Sn]=3×10^{17}	4.4×10^{11}	7.95×10^{16}	550 °C, 10 min
(12)	NTD:[Sn]=1×10^{18}	4.7×10^{11}	2.65×10^{17}	550 °C, 10 min
(13)	Conven. Fe-Doped [Fe]=$(3–4)\times10^{16}$			
(14)	Conven. Sn-Doped [Sn]=4.8×10^{17}			

from 2×10^{15} to 2×10^{18} cm^{-3}. Due to the low neutron flux used ($\phi\leqslant10^{12}$ n/cm s), the samples remained at room temperature during the irradiation. The thermal neutron flux and fluences as well as the expected Sn concentrations are listed in Table I.

Apart from the irradiated and the as-grown samples, two others InP samples were measured: Conventionally Fe and Sn doped with a free carrier concentration at room temperature of about 10^8 cm^{-3} and 4.8×10^{17} cm^{-3}, respectively.

The thermal annealing were performed at 550 °C during 10 min under an argon gas flow at ambient pressure.

A conventional fast–fast lifetime equipment with two BaF_2 detectors has been used. The time resolution [full width at half-maximum (FWHM)] was 250 ps for ^{60}Co and the time calibration was 18.7 ps/channel. The source, ^{22}NaCl (about 10 μ Ci) sealed in a kapton foil (1.42 g/cm^3), is sandwiched between two samples. Positron lifetime spectra were recorded at room temperature and the data were analyzed with POSITRONFIT.[13,14]

The lifetime spectra contained no less than 2×10^6 counts and a source correction with two components was used in the analysis: The first one of 382 ps with 12% intensity (kapton foil contribution), and the second one of 800 ps with 1% intensity. Each measurement was repeated at least three times.

III. RESULTS AND DISCUSSION

The time spectrum of the undoped sample (sample 1) has been fitted by one exponential decay component. The obtained value (246±2 ps) agrees with those previously reported by Dlubek *et al.*[7] and Puska *et al.*[15] for this material. A two terms fit, as suggested by other authors (Bretagnon *et al.*[8]), yields a very low value for I_2 (<1%) and does not substantially improve the variance of the fit. The lifetime values found for n-type (Sn-doped) and semi-insulating (Fe-

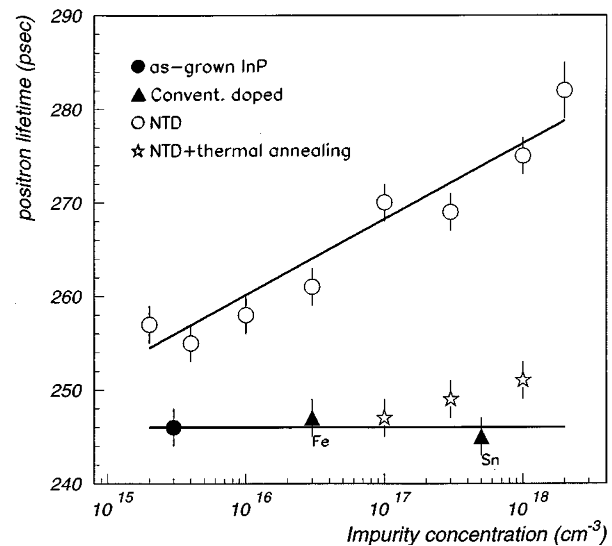

FIG. 1. Lifetime parameters versus the Impurity concentration. Empty symbols—correspond to samples in the as-irradiated condition. Triangles—Crystals conventionally doped with Fe (3×10^{16} cm^{-3}) and Sn (4.8×10^{17} cm^{-3}). Full circle—As-grown material. Stars—Samples annealed at 550 °C during 10 min and then measured at room temperature. The horizontal dashed line represents the mean value obtained in undoped and conventionally doped samples.

doped) samples were 245±2 and 247±2 ps, respectively. These results are closed to those obtained in the undoped samples.

The results of the experiments have been represented in Fig. 1. Empty symbols correspond to samples in the as-irradiated condition and the measures are plotted at their Sn-expected concentrations. Full symbols correspond to the as-grown and conventionally doped samples. In the x axis the impurity concentration has been represented. Error bars are include when they are larger than the symbol size.

The full circle corresponds to the as-grown material used as starting material for the neutron transmutation doping experiments. This material was n type with n(300 K)~$(2–4)\times10^{15}$ cm^{-3}. The room temperature electron concentration comes from the existence of residual native donors in the as-grown material. The measured lifetime value is 246 ps. The semi-insulating sample (Fe-doped) had a resistivity value in the range of $\sim10^7$ Ω cm. The semi-insulating behavior is due to the presence of Fe atoms which act as acceptors and compensate the residual donors. The [Fe] value is about 3×10^{16} cm^{-3} and the lifetime value is 247±2 ps. The Sn-doped sample is n type with an electron concentration measured by Hall effect of n(300 K)~4.8×10^{17} cm^{-3}. The lifetime value found is 245±2 ps.

The horizontal solid line represents the average value of lifetime obtained in undoped and conventionally doped InP samples. It can be conclude that the nature and concentration of dopants does not affect the positron annihilation in doped samples, in agreement with the results reported by Bretagnon *et al.*[8] for InP with different types of dopants (Fe, Zn, S, Si) and carrier concentrations. This result means that the positron lifetime in InP does not depend of the Fermi level position. Unlike, in other III–V materials, as for example GaAs, the lifetime seems to be a function of the Fermi

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level.[16] This difference might be due to the fact that in GaAs the Fermi level is controlled by the midgap level EL2. This level is an intrinsic defect which can trap positrons depending on it state of charge.

The open circles in Fig. 1 correspond to the irradiated samples. In this case, the x axis represents the expected Sn concentration calculated from the irradiation dose. The tin concentration can be written as[9]

$$[\mathrm{Sn}]=(\phi t)\sum_i n_i\sigma_c^i \tag{2}$$

where (ϕt) is the thermal neutron fluence, and n_i and σ_c^i are the concentration and capture cross-section of the In ith isotope, respectively. For InP, the neutron absorption coefficient, $\Sigma_i n_i \sigma_c^i$, reaches a value of 3.77 cm^{-1}.

The lifetime spectra were analyzed with one component. It is found that the neutron irradiated samples yielded a lifetime longer than the measured one on undoped InP. The lifetime values show a monotonous increase with the thermal neutron dose and vary from 257 ± 2 ps (2×10^{15} cm^{-3}) to 282 ± 3 ps (2×10^{18} cm^{-3}). This effect shows that certain lattice defects induced by thermal neutrons act as effective positron traps.

In our case, the lifetime obtained from the single-component analyses should be interpreted as a weighted mean lifetime of positrons annihilating in the bulk and at the induced defects. According to the positron trapping model this mean lifetime parameter, $\bar{\tau}$, is a linear superposition of their values in the bulk, τ_b, and at the positron traps, τ_d:

$$\bar{\tau}=(1-\alpha_d)\tau_b+\alpha_d\tau_d, \tag{3}$$

where α_d is the fraction of positrons annihilating at vacancy defects. These defects should be produced by the γ and β^- particles involved in the doping process. In this process, vacancies can be formed in both sublattices. The positrons can be trapped by one or both of them and for that, τ_d would be a mixture of two contributions: annihilations in $V_{\mathrm{In}}-\mathrm{Sn}_{\mathrm{In}}$ (In vacancy-Sn interstitial) and in V_P-S_{In} (P vacancy-S interstitial).

Theoretical calculations[15] suggest that the increase in lifetime from the bulk lifetime is 50 ps for V_{In} and 28 ps for V_P. Experimentally, we obtain an increase in $\bar{\tau}$ relative to the bulk lifetime close to 40 ps ($[\mathrm{Sn}]=2\times10^{18}$ cm^{-3}). By comparing both results, we can conclude that the increase observed in $\bar{\tau}$ is more probably due to positrons trapped at Indium than at P vacancy related defects. This explanation agrees well with the type of defects expected from the transmutation process where mainly points defects due to the γ and β^- emissions should be generated. During the transmutation reaction of ^{115}In atoms, the average of the recoil energy transmitted to the involved atom through the β^- decay is close to 15 eV which is enough to create vacancy-interstitial pairs in the In sublattice since only 5–8 eV is required for the creation of Frenkel defects in III–V materials. Furthermore, the β^- decay is always accompanied by the emission of a cascade of 4 or 5 γ rays. The induced γ recoils of the ^{116}In (excited nuclei) occur with an average energy of 50 eV,[17] which confirms that the transmuted nuclei are come to rest not far away from their initial positions.

As the neutron absorption coefficient is 1000 times higher for In isotopes than for the P nucleus, the probability to create a vacancy-related defect in the In-sublattice should be 1000 times higher than those created in the P sublattice.

Concerning the defects create by the fast neutrons, it is expected to be similar to those created by ion implantation which consist mainly in clusters of about 50–100 defects.[12] However, as the rapid neutron rate and the capture cross-section at this energies are low (~b), one finds that the number of defects due to the β recoil is very large compared with those created by the fast neutrons.

The three heaviest irradiated samples (10, 11, and 12) were annealed at 550 °C during 10 min and then measured at room temperature. Again, the spectra are well fitted with only one component. The lifetime values obtained for these samples have been plotted in Fig. 1.

The decrease observed in the lifetime shows that most of the positron traps produced during the transmutation process have been effectively annealed at this temperature.

As it can be seen in Fig. 1, the lifetime values for the annealed samples are slightly longer than the measured one on as-grown InP (246 ± 2 ps) indicating the present of some remaining defects. This result can not be associated with the annealing time used (10 min) because electrical measurements show that the samples exhibit the same electron concentration for different annealing times (10 s, 10 min or 30 s) at 550 °C.[9] The remaining defects are probably neutral since these only affect the low temperature mobility value and do not affect the carrier concentration.[10]

IV. CONCLUSIONS

The evolution of the positron lifetime in NTD-InP crystals is presented. $\bar{\tau}$ shows a monotonous increase with the neutron dose. This effect shows that certain lattice defects, induced by thermal neutrons, act as effective positron traps. These defects are identified as indium vacancies impurities.

From thermal-annealing experiments at 550 °C, a decrease of mean positron-lifetime is observed. In the samples doped above $[\mathrm{Sn}]=10^{17}$ cm^{-3}, τ_b is not completely recovered suggesting that certain neutral defects remain in the crystal lattice.

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