

Positron Lifetime Spectroscopy

The positron lifetime τ is a function of the electron density at the annihilation site. The annihilation rate λ , which is the reciprocal of the positron lifetime τ , is given by the overlap of the positron density $n_+(\mathbf{r}) = |\psi^+(\mathbf{r})|^2$ and the electron density $n_-(\mathbf{r})$ (Nieminen and Manninen 1979),

$$\lambda = \frac{1}{\tau} = \pi r_0^2 c \int |\psi^+(\mathbf{r})|^2 n_-(\mathbf{r}) \gamma d\mathbf{r}. \quad (1)$$

r_0 is the classical electron radius, c the speed of light, and \mathbf{r} the position vector. The correlation function $\gamma = \gamma[n_-(\mathbf{r})] = 1 + \Delta n_-/n_-$ describes the increase Δn_- in the electron density due to the Coulomb attraction between a positron and an electron. This effect is called enhancement.

When positrons are trapped in open-volume defects, such as in vacancies and their agglomerates, the positron lifetime increases with respect to the defect-free sample. This is due to the locally reduced electron density of the defect. Thus, a longer lifetime component, which is a measure of the size of the open volume, appears. The strength of this component, i.e. its intensity, is directly related to the defect concentration. In principle, both items of information, i.e. the kind and concentration of the defect under investigation, can be obtained independently by a single measurement. This is the major advantage of positron lifetime spectroscopy compared with angular correlation of annihilation radiation or Doppler-broadening spectroscopy with respect to defect issues.

1. Basics of the Measurement

The conventional positron lifetime measurement is possible since a γ -quantum with an energy of 1.27 MeV is emitted almost simultaneously with the positron in the ^{22}Na source. The positron energy, which extends up to 540 keV, decreases in the sample within a few picoseconds by non-elastic interactions. The mean positron penetration depth of this so-called thermalization process is of the order of 100 μm . The thermalization time usually amounts to a few picoseconds. It is thus small compared with the positron lifetime and can be neglected. On reaching thermal energies, the positron diffuses in the periodic lattice potential before it is possibly trapped in a lattice defect. The diffusion length is in of order of 100 nm. This distance determines the number of atoms to be probed for positron traps during the positron lifetime. Hence, the diffusion length strongly determines the sensitivity of the positron methods to detect defects.

The positron lifetime of a single event can be measured by detecting the time difference between the birth γ -quantum of the β^+ -decay in the source and one of the annihilation γ -quanta of an energy of 511 keV. The activity of the source must be sufficiently low in order to ensure that on average only one positron is in the sample. This avoids the intermixing of start and stop quanta originating from different annihilation events. A special "sandwich" arrangement of foil source, samples, and detectors guarantees that all positrons emitted from the source are penetrating the sample material. The γ -rays are converted by scintillator-photomultiplier detectors into analog electrical pulses. The pulses are processed by discriminators. Their output pulses start and stop a time-to-amplitude converter as an "electronic stopwatch". The amplitude of the output pulse is proportional to the time difference between the birth and the annihilation γ -quanta and, thus, represents a measure of the positron lifetime. The single annihilation event is stored after analog-digital conversion in the memory of a multi-channel analyzer. The channel numbers represent the time scale. In order to obtain the complete lifetime spectrum, more than 10^6 annihilation events must be recorded.

The scheme of the positron lifetime measurement is shown in Fig 1. BaF₂ or plastic scintillators and photomultipliers with a short pulse rise-time are used to obtain a high time resolution. The discriminators suppress noise and generate standard timing pulses by the constant-fraction discrimination principle. This principle is favored over leading-edge discrimination in order to ensure stable time markers independent of the pulse height. Another task is to guarantee that the 1.27-MeV and 0.51-MeV quanta are accepted only in the appropriate channels. The discriminators are of differential type (single-channel analyzer) and accept input pulses within an adjustable energy window.

The timing pulses are used to start and stop the charging of a capacitor in the time-to-amplitude converter (TAC). The time linearity is ensured there by constant-current charging that is stopped at the arrival of the stop pulse originating from the annihilation γ -quantum. The stop pulse is coax-cable delayed in order to shift the time spectrum into a linear region of the TAC. The spectrum is stored in a multi-channel analyzer.

This experimental arrangement is called “fast–fast coincidence” setup. The term is related to the fact that the time measurement as well as the energy selection is performed in a fast channel. A slow channel was used for energy selection when fast differential discriminators were not available at the beginning of positron lifetime experiments. This arrangement is called a fast–slow setup.

Inexpensive multi-channel plug-in boards for personal computers with about 2000 channels are sufficient for storing the spectra. The time resolution of the spectrometer is determined mainly by the scintillator–multiplier part and ranges between 180 and 280 ps. The practical consequence of this relatively poor resolution is the limitation of the determination of positron lifetime components larger than about 50 ps. The determination of positron lifetimes can, however, be carried out with an accuracy of about 1 ps.

2. Data Treatment

The time-dependent positron decay spectrum $D(t)$ in the sample is given by

$$D(t) = \sum_{i=1}^{k+1} I_i \exp\left(-\frac{t}{\tau_i}\right). \quad (2)$$

k different defect types contributing to the positron trapping are related to $k + 1$ components in the spectra with the individual lifetimes τ_i and intensities I_i . If no positron traps are present in the sample, (2) is reduced to $D(t) = \exp(-t/\tau_b)$, where τ_b is the positron lifetime in the defect-free bulk of the sample. The positron lifetime spectrum $N(t)$ is the absolute value of the time derivative of the positron decay spectrum $D(t)$,

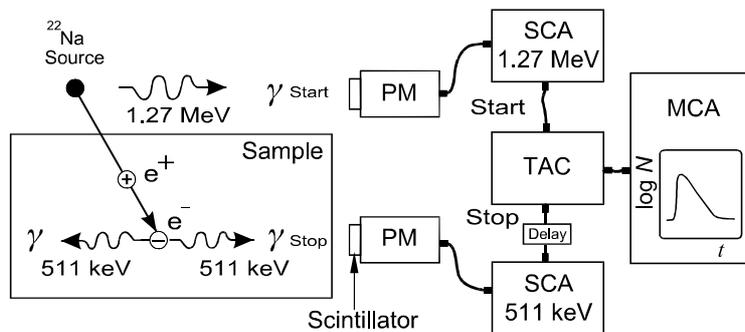


Fig. 1. Scheme of the positron lifetime experiment in fast–fast coincidence. The lifetime is measured as the time difference between the appearance of the start and stop γ -quanta (PM—photomultiplier, SCA—single-channel analyzer). The amplitude of the time-to-amplitude converter (TAC) analog output pulse is proportional to this time difference. The whole lifetime spectrum $N(t)$ is stored in a multi-channel analyzer (MCA).

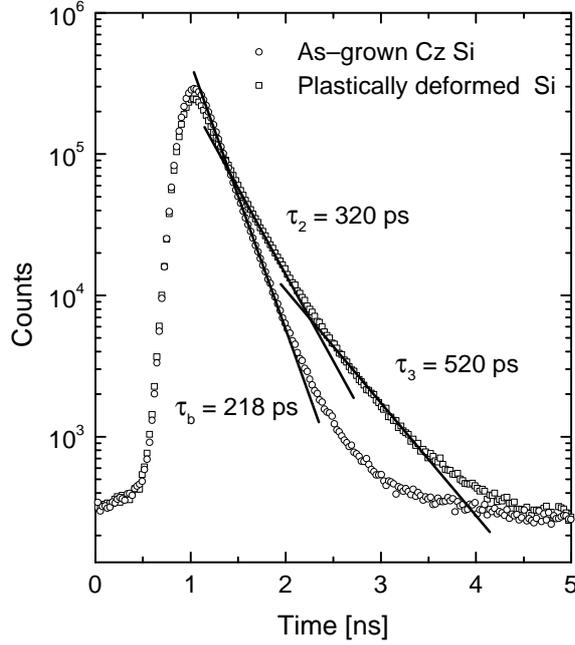


Fig. 2. Experimental positron lifetime spectra obtained in as-grown and in plastically deformed Czochralski-grown (Cz) silicon (Hübner et al. 1997b). The curve of the deformed sample (3 % strain, deformation temperature 775 °C), is located significantly higher, indicating the occurrence of long-lived lifetime components. After the decomposition of the upper spectrum, the obtained lifetime components τ_2 and τ_3 are added as straight lines in the semi-logarithmic plot for illustration. The τ_1 component is not indicated as a straight line ($\tau_1 = 120$ ps). Only one lifetime component corresponding to the positron bulk lifetime τ_b is found in the as-grown sample. The deviations from the straight line at higher times are due to annihilations in the source and the background contribution. The Gaussian-like shape of the left part of the curve is mainly caused by the resolution function.

$$N(t) = \sum_{i=1}^{k+1} \frac{I_i}{\tau_i} \exp\left(-\frac{t}{\tau_i}\right). \quad (3)$$

The spectrum is shifted on the time scale by the delay cable (Fig. 1) to time zero t_0 , and consequently the time t in (2) and (3) has to be replaced by $t - t_0$.

Typical positron lifetime spectra obtained in as-grown silicon and in Si after plastic deformation are shown in Fig. 2. The experimentally obtained spectra differ from the analytical description (3) mainly by the convolution with the time resolution function $F_t(t)$. In general, it is approximated by a sum of several Gaussians or one Gaussian with exponential tails. In the case of the use of plastic scintillators, it is often sufficient to take a single Gaussian $G(t)$ centered at t_0 ,

$$G(t) = \frac{1}{\sigma\sqrt{\pi}} \exp\left[-\left(\frac{t-t_0}{\sigma}\right)^2\right]. \quad (4)$$

The time resolution is characterized by the full width at half maximum, FWHM,

$$\text{FWHM} = 2\sigma_s \sqrt{\ln 2}. \quad (5)$$

with the standard deviation σ_s . The convolution of the spectra (2) with $F_t(t)$ gives the convolved decay spectrum

$$D_f(t) = \int_{-\infty}^{\infty} D(t-t') F_t(t') dt'. \quad (6)$$

If $F_i(t) = G(t)$, i.e. a single Gaussian is taken as the resolution function, the positron decay spectrum is given as

$$D_f(t) = \sum_{i=1}^{k+1} \frac{I_i}{2} \exp\left[-\frac{t - t_0 - \sigma_s^2 / (4\tau_i)}{\tau_i}\right] \left[1 - \operatorname{erf}\left(\frac{1}{2\sigma_s \tau_i} - \frac{t - t_0}{\sigma_s}\right)\right]. \quad (6)$$

Standard computer programs based on Gauss–Newton non-linear fitting routines are available for the decomposition of the spectra (e.g. LIFSPECFIT by Puska 1978, POSITRONFIT or PATFIT by Kirkegaard et al. 1989). A model spectrum with a given number of decay components and a given resolution function is used for the least squares fit to the measured spectrum by the variation of the parameters of the lifetime components. The linear parameters, i.e. the intensities and the background, are fitted independently of the non-linear ones, which are the lifetimes τ_i and the time-zero channel t_0 .

Only a one-component lifetime spectrum is obtained in a defect-free semiconductor. The deformed sample of Fig. 2 contains two defect types with open volume, viz. vacancies and voids, and thus, three distinct components appear as a superposition of straight lines in the semi-logarithmic plot, which are to be folded by the time resolution function to give the lifetime spectrum. The spectra additionally contain background contributions as well as annihilation events in the positron source.

The source correction has to be performed after background subtraction. This means subtracting the characteristic lifetime spectrum of the source. The determination of this source spectrum is rather complicated and usually carried out in such a way that a one-component spectrum of a defect-free sample is analyzed. The source components are varied using a single-component fit to get the best fit, i.e. the smallest variance. The problem of the source correction was investigated in detail on measured and simulated spectra (Staab et al. 1996). It was found that sources prepared from Al foils provide a three-component source spectrum. The fraction of the annihilations in the source is not only determined by the source itself but it is also a function of the atomic number of the sample and increases due to multiple positron backscattering through the source.

The determination of the number of components in samples with unknown defect populations is started by a one-component fit of the spectrum. Additional components are added as long as the variance of the fit decreases. However, the possibility of obtaining a reliable multi-component fit is restricted due to the limited time resolution of the spectrometer and the statistics of the measurements. A number of one to four components can be resolved, depending on the intensity and the mutual separation of the individual lifetime components, the time resolution, and the statistics. These effects were studied with Monte-Carlo-simulated as well as experimentally obtained spectra (Somieski et al. 1996). It was found that the possibility of decomposing the positron lifetime spectra depends essentially on the number of components in the spectrum, the distance of the individual lifetimes, and the statistics of the measurement. At least five million events should be collected for a reliable lifetime separation, even if only two components are present. If only the average positron lifetime is to be measured, a lower number of counts is sufficient.

The decomposition of spectra described above is related to spectra having discrete lifetime components, as is expected in most cases for semiconductors. A nearly continuous distribution of lifetime components, as found in polymers (Jean 1995), may occur in semiconductors if vacancy clusters of different sizes are present. In this case, and if the number of components is too high to be resolved individually, a continuous distribution of lifetimes can be fitted to the experimental data and an intensity versus lifetime plot is obtained. Such a data analysis is based on Laplace transformation of the measured spectrum (CONTIN program, Provencher 1982).

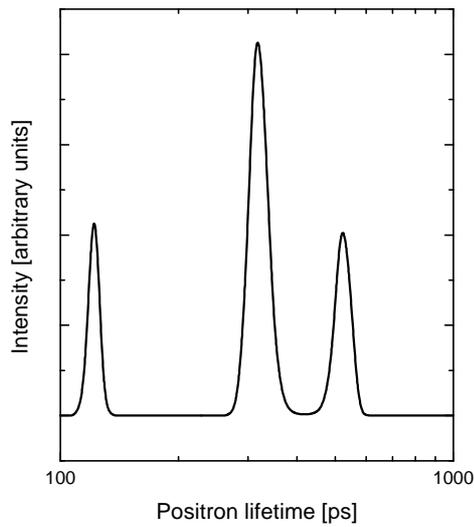


Fig. 3. Result of a maximum entropy of lifetime (MELT) analysis of the positron lifetimes of plastically deformed Czochralski-grown silicon (Hübner et al. 1997b). The sample is the same as in Fig. 2 (3.5 % deformation at 775 °C). The peaks obtained correspond well to the discrete lifetimes from the decomposition of the spectrum in Fig. 2, $\tau_1 = 120$ ps, $\tau_2 = 320$ ps, and $\tau_3 = 520$ ps.

Another approach has been made by Shukla et al. (1993) using linear filtering and the method of maximum entropy (MELT program). Principally, both methods can also be applied to discrete lifetime spectra. The output is a graph displaying the intensity versus lifetime. Knowledge of the number of components is not required. This is an advantage for multi-component spectra, but it has not found a broad application to semiconductors up to now. The result of a MELT¹ analysis of a plastically deformed silicon sample is shown in Fig. 3. The same lifetimes were obtained from the decomposition by a non-linear fit according to (3) taking into account the resolution function.

¹ Maximum entropy of lifetime.

