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Positron Lifetimes of the 2H-TASE2 Crystal as a Function of Temperature

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POSITRON LIFETIMES OF THE 2H-TASE2 CRYSTAL AS A FUNCTION OF TEMPERATURE

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The positron lifetimes of a 2H-TaSe₂ crystal were measured as a function of temperature between 10 and 490 K. Two distinct and constant lifetimes, i.e. $0.130 \pm .007$ and $.378 \pm .008$ nsec, were found in the temperature range studied. These two lifetimes are thought due to the anisotropy character of the two-dimensional layered structure of the crystal. The applications of the positron annihilation methods to study the charge density wave and related phenomena are discussed.

INTRODUCTION

Most of crystal compounds form regular periodical lattices except for the imperfections, impurities or boundaries. However, it has been thought some times that a few compounds, their regular lattices become distorted in the absence of the internal impurities or the external deformations. The ideas of the charge density wave (CDW) that displays a long modulation of the charge density in the lattices were first considered by Peierls and Frohlich¹ several years ago. The interests of CDW and related physical anomalies have grown enormously because a few ideal low dimensional crystals became available recently.

Neutron and x-ray diffraction methods offer the most convincing evidences on the lattice distortion and the structural transformations of the CDW compounds.² Most of modern

techniques for physical measurements such as resistivity, susceptibility, thermal conductivity, electron diffraction, NMR, IR, and Raman scattering, etc. have been applied to study the CDW systems. On the other hand, the positron annihilation method³, which has been known as one of the most powerful techniques to study the imperfections and the phase transformations of crystals. And yet it has not been applied to study the CDW systems.

In this paper, we chose a TaSe₂ crystal as our first system to study the CDW and related phenomena by the positron annihilation method. The 2H-TaSe₂ crystal has been reported recently⁴, at least three CDW phase transitions at 93K, 102K, and 112K. A reasonable size of the single crystal is also available. Therefore, we report the first attempt of the positron lifetimes of this crystal between 10 and 490K to search for the applications of positrons to the low dimensional crystals.

EXPERIMENTAL

Two pieces of 2H-TaSe₂ single crystals (thickness 3 mm and about 5mm X 5mm) were grown as previously described.⁵ The positron sources (⁵⁴Co Na²²) were directly deposited on the surface of the crystal and sandwiched in between crystals. The sandwiched crystals were annealed at 589K for 48 hours under the Ar atmosphere. The positron lifetime spectrometer is shown in Figure 1. The decay of Na²² emits a positron and a 1.28 Mev photon nearly simultaneously. This 1.28 Mev photon

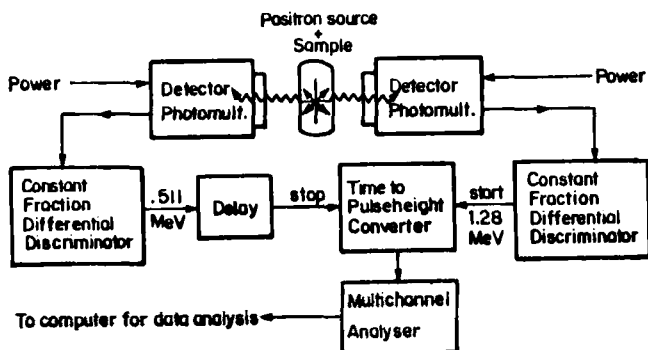


FIGURE 1 A positron lifetime spectrometer at the University of Missouri-Kansas City.

serves as a starting signal and the annihilated photon (.511 Mev) serves as a stopping signal for the lifetime measurement. The resolution of the lifetime spectrometer was found 270 psec by measuring the coincident photons from a Co-60 γ -source. The obtained lifetime spectra were resolved into a multi-exponential function by a computer program - POSITRONFIT.⁶ A typical positron lifetime spectrum is shown in the Figure 2.

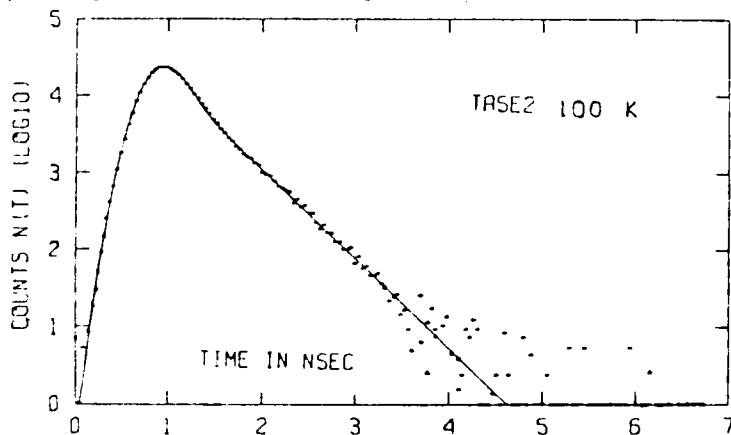


FIGURE 2 A typical lifetime spectrum of a 2H-Tase2 crystal. The line is the computer fitted result in two-exponential function.

The positron source contribution to the lifetime spectra was estimated less than 2% and was found insignificant to the lifetime results. Every lifetime spectrum was carefully resolved into one-, two- and three-components and only two-component results were found consistently good χ^2 fit from the computer program. Therefore we believe only two positron lifetimes existed in the 2H-Tase2 crystals at the temperature range studied here. The low temperature measurements were made at a cryostat under the He-atmosphere. The temperature deviation was less than ± 0.2 K. The high temperature (>300 K) measurements were made at a furnace under the Ar atmosphere. The temperatures were controlled better than 1 K.

RESULTS AND DISCUSSIONS

When positrons are emitted from nuclear decay, they are quickly thermalized from energetic states in the lattice in the time less than 10^{-12} sec. These thermalized positrons display Bloch waves along the perfect crystals. The positron lifetime in

the crystal depends on the overlap between the positronic and electronic wave functions in the same site. In the well-annealed crystals, the observed positron lifetime is inversely proportional to the electronic density of the crystals.³ For a two-dimensional layered compound, there exists two distinct electronic distributions, along the layered plane and perpendicular to the plane. The conductivity parallel to the layer is larger than the perpendicular conductivity.² We expect to observe at least two distinct positron lifetimes in this two dimensional crystal.

The results of the lifetime in two components are shown in Figure 3. The short lifetime component, $\sim .13$ nsec, is assigned to the positrons annihilation parallel to the layer and the long-lived component, $\sim .378$ nsec, perpendicular to the layer. The higher conductivity parallel to the layer, thus a shorter lifetime is observed. Both two lifetimes are almost constant between 10 to 320K. The intensities of the long-lived component show similar results in Figure 4.

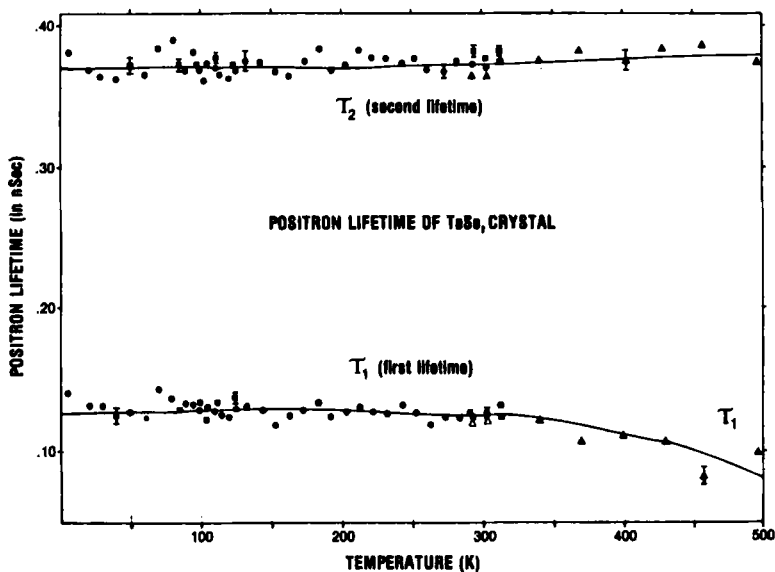


FIGURE 3 Positron lifetimes of the 2H-TaSe₂ as a function of temperature. The line is drawn for eye-guide only.

The 2H-TaSe₂ crystals form a two-dimensional CDW along the layered plane. Three CDW phase transitions were observed at 93K, 112K and 122K.⁴ We have concentrated the measurements between 90K and 130K as shown in Figure 3 and 4. The results

obviously indicate that positron lifetimes do not change with the CDW phase transitions in 2H-Tase2 crystals. The slightly changes of positron lifetimes and the long-lived components above 300K are due to the vacancy formation by the thermal process. If positron lifetimes are sensitive to CDW phase transitions, we should have observed the change of short lifetimes near 100K. The lack of the response of positron lifetimes to these CDW transitions could be due to the fact that positronic wavelength in the lattice near 100K ($\sim 100\text{\AA}$) is not long enough to detect the charge density wave modulations. Positrons are probably still too localized compared to the charge density wave modulations. On the other hand, we also could not rule out the possibility that the intrinsic defects in the lattice could have also blocked the motions of positrons in the lattice. The crystals were decomposed before we could heat crystals up to the well-annealed temperatures.

Two other positron annihilation techniques, i.e. Doppler broadening and the angular correlation of the annihilated photons could measure the momentum distribution functions in the lattice. These are useful methods to detect Fermi instability due to CDW. The experimental sensitivities of these methods are also better than the lifetime methods. These researches are in the progress at our laboratory.

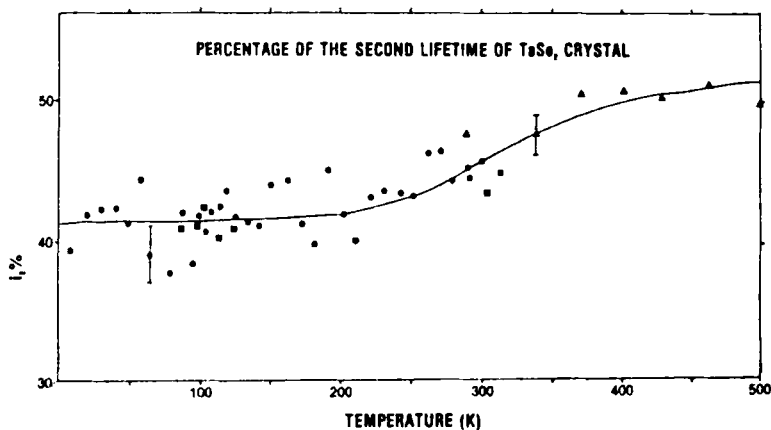


FIGURE 4 Long-lived components from positron lifetimes spectrum of the 2H-Tase2 crystal as a function of temperature. The line is drawn for the eye-guide only.

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