POSITRON MEASUREMENTS IN 2H-$TaSe_2$ CRYSTALS*

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CONF-850169--10

DE85 010507

JANUARY 1985

*This work is partially supported by the U.S. Department of Energy.

Submitted by the University of Missouri-Kansas City, and presented at the Seventh International Conference on Positron Annihilation, New Delhi, India, 6-11 January 1985.

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Temperature-dependent positron annihilation lifetime and Doppler broadening experiments are reported on single crystals of $2H-TaSe_2$ to search for effects from known charge-density-wave (CDW) phase transitions. The positron lifetime in the perfect lattice and in positron trapping sites were found to be 0.173 and 0.378 ns respectively. The apparent activation energy for the thermally generated trapping sites was found to be 0.12 eV. Doppler broadening spectra exhibited no response to the known CDW phase transitions, nor any significant overall anisotropy in their temperature dependence.

1. INTRODUCTION

A charge density wave (CDW) is a periodic spatial oscillation of the free-electron charge density in conducting materials.[1] Important physical properties of solids, such as superconductivity, electric resistivity, magnetic susceptibility and thermal conductivity, all exhibit responses which are directly associated with the CDW phenomena.

Recently, interest in using positron annihilation to gain insight about the Fermi surface shape of layered and two dimensional materials has been increased. Boileau and coworkers[2] have reported detailed Doppler, 1D-ACAR, and lifetime results for $1T-TaS_2$ that appear to indicate that the idea of positrons trapped in states associated with CDWs may be valid, as well as providing some preliminary experimental insight of the Fermi surface using the Lock-Crisp-West (LCW) theorem. We wish to report our results on positron lifetime and Doppler broadening measurements for an analogous system, $2H-TaSe_2$.[3]

2. EXPERIMENTAL

Single crystals of $2H-TaSe_2$ were grown by a direct chemical reaction between Ta and Se elements as described[4] earlier with regard to neutron diffraction experiments. The positron source (Na^{22}) was sandwiched between two identical crystals (2mm x 5mm x 5mm) and then was annealed at 785K for 48 hrs. Positron lifetime spectra were measured by using a conventional fast-fast coincident technique and then they were fit by using the computer program POSITRONFIT EXTENDED. A Ge(Li) detector (resolution 1.6 KeV at 497 KeV) was used for the Doppler broadening experiments.

3. RESULTS AND DISCUSSION:

(1) POSITRON LIFETIME OF $2H-TaSe_2$:

The lifetime results for $2H-TaSe_2$ crystals between 10 K and 490 K have been reported.[5] Two constant lifetimes were observed, i.e. 0.130 ± .007 and 0.378 ± .008 ns between 10 K and 300 K. The intensity of the long-lived component was 42% between 10 K and 300 K and was found to be temperature independent. The positron lifetime results between 300 K and 787 K are shown in Fig. 1.

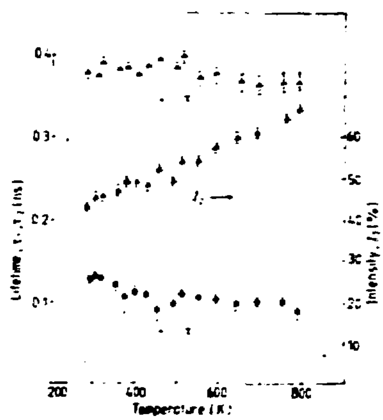

Fig. 1. Positron lifetime in $2H-TaSe_2$

We interpret the variation of lifetime results in terms of a two-state trapping model with the following expressions:

$$\frac{1}{\lambda_1} = \frac{1}{(\lambda_B + k)} \qquad (1)$$

$$\frac{1}{\lambda_2} = \frac{1}{\lambda_T} \quad (2)$$

$$I_2 = k/(\lambda_B - \lambda_T - k) \quad (3)$$

λ_B = positron annihilation rate in the Bloch state,
λ_T = positron annihilation rate in the defect state,
k = positron trapping rate of Bloch state to defects.
λ_1 = observed short-lived positron decay rate,
λ_2 = observed long-lived positron decay rate.
I_2 = intensity of long-lived component.

The validity of the trapping model has been tested by observing a nearly constant value of λ_B with temperature according to the following equation:

$$\lambda_B = \lambda_1 I_1 + \lambda_2 I_2 \quad (4)$$

The calculated results of λ_B are plotted in Fig. 2.

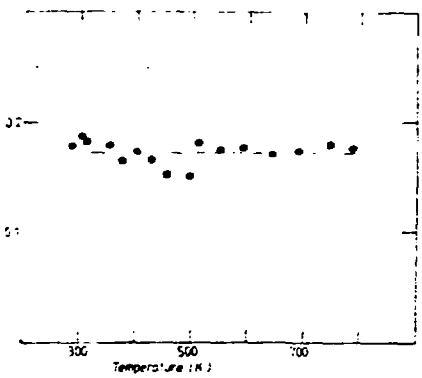

Fig. 2. The positron lifetime in the bulk of $TaSe_2$.

In order to obtain the trapping rates and related activation energy, we modify the trapping model since the crystal starts with extrinsic defects which compete for the positron along with the thermal equilibrium defect population. We employ the following relationship:

$$k = k_v + k_e$$

$$k_e = I_2^o (\lambda_1^o - \lambda_2^o)$$

$$k = I_2(\lambda_1(T) - \lambda_2(T))$$

k = overall trapping rate,
k_v = trapping rate in thermal defects.
k_e = trapping rate in extrinsic defects.

The superscripts o refer to the states in extrinsic defects. A plot of $\ln k_v$ vs. 1/T gives an activation energy of 0.12 ± 0.03 eV as shown in Fig. 3.

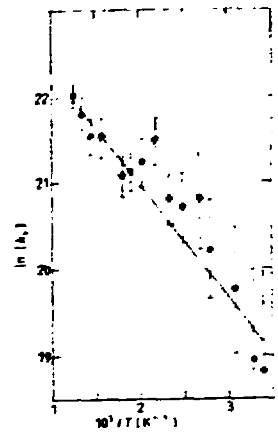

Fig. 3. Arrhenius plot of k_v in $TaSe_2$.

The most likely candidate for this activation energy is some type of vacancy formation enthalpy. We have recently measured the positron lifetimes of γ-irradiated $TaSe_2$ crystals[7] and found a similar vacancy trapped by positrons.

(2) DOPPLER BROADENING SPECTRA OF $2H\text{-}TaSe_2$

The results of the present investigation of $2H\text{-}TaSe_2$ for the Doppler broadening line shape parameters, S(peak) and W(wing) are shown in Fig. 4. Two crystallographic orientations were chosen, the c-axis parallel to the γ-ray detector and the a-axis parallel to the detector. The results of Fig. 4 show no significant change in line shape parameter near the known CDW transition temperatures[6] of 93 K, 112 K and 122 K. This negative finding contrasts with the positive finding of Boileau et. al. on the analogous $1T\text{-}TaS_2$ crystals. We observed the maximum change of the parameters S or W to be less than the experimental sensitivity, limit of 0.1% for the present Doppler broadening technique.

One reason for this negative finding in responding to CDW transitions for the $2H\text{-}TaSe_2$ system is the small change in lattice distortion coupled with the CDW transitions as compared with the $1T\text{-}TaS_2$ system. The maximum change of the lattice parameter associated with $2H\text{-}TaSe_2$ is only 0.0016% along the c-axis while the change for $1T\text{-}TaS_2$ is 0.46%.

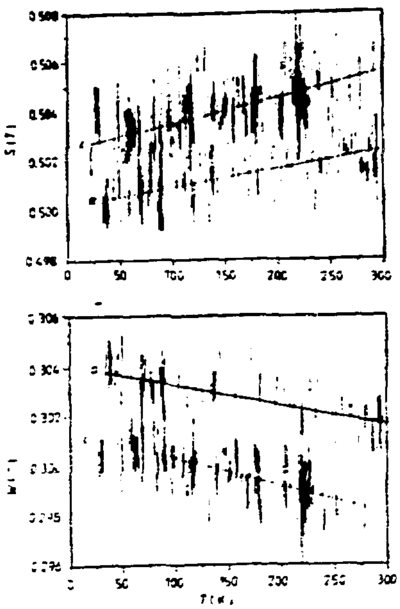

Fig. 4. Peak (S) and wing (W) parameters from Doppler broadening spectra in $TaSe_2$. c refers to the c-axis parallel to the detector and a refers to the a-axis parallel to the detector.

4. CONCLUSION

(1) The detection of the CDW transition by the Doppler broadening technique is limited to lattice distortion of >0.01%.

(2) Negative results in detecting CDW transitions for $2H\text{-}TaSe_2$ are likely a consequence of the small change in lattice paramters.

(3) Positron lifetime results yield a positron trapping site formation enthalpy for $2H\text{-}TaSe_2$ to be 0.12 eV (unusually small compared to vacancy formation enthalpy in metals).

+ Work is partially supported by US DOE under contract no. W-31-109 Eng-38.

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