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Positron annihilation study in SmFeAsO and $SmFeAsO_{0.82}F_{0.18}$

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$SmFeAsO_{1-x}F_x$ polycrystalline samples were first studied by positron annihilation lifetime spectroscopy and Doppler-broadening spectroscopy, combined with the calculated results of positron lifetime. The experimental results agree well with the calculated positron bulk lifetime in SmFeAsO and SmFeAsF crystals. The temperature dependence of S-parameter shows a remarkable difference between the parent and superconductor. An abrupt jump of S-parameter is detected around 150 K in parent sample, however, two different slopes were shown below and above superconducting transition. The linearity S-T plot determines one-type of defects through the superconducting transition which must play an important role in superconductivity. © *2010 American Institute of Physics*. [doi:10.1063/1.3293299]

Recently, a new high temperature superconductor $SmFeAsO_xF_{1-x}$ (Refs. 1 and 2) with a ZrCuSiAs-type structure was discovered. It is a layered rare-earth metal oxypnictides LnMPnO (Ln=La, Pr, Ce, and Sm; M=Fe, Co, Ni, and Ru; Pn=P and As). Before it was discovered, only in the copper oxide superconductor has been obtained a T_c higher than 40 K.[3,4] This discovery provided a new material base for studying the origin of the electron pairing and superconductivity.

It is of considerable interest to study the role of defects and the electronic momentum distribution, particularly when the temperature is close to T_c. Positron annihilation techniques[5] are considered as a unique probe nondestructive with a sensitivity of 10^{-6} (ppm) order, with which the vacancy concentration can be directly obtained.[6,7] Positron annihilation techniques have been used widely in the research of high-T_c copper oxide superconductors.[8–10] In this letter, positron annihilation lifetime spectroscopy (PALS) measurements of SmFeAsO and $SmFeAsO_{0.82}F_{0.18}$ samples were first carried out at 298 K. Superposed-neutral-atom (SNA) model and finite-difference method (FD) are used to calculate the positron lifetime of SmFeAsO and SmFeAsF crystals with different types of vacancies. Furthermore, we have employed Doppler-broadening spectroscopy (DBS) and helium cryostat equipment to study the iron-base high-T_c superconductors and reported the change of S parameter as the function of temperature.

The $SmFeAsO_{0.82}F_{0.18}$ (T_c=48.5 K) sample was synthesized by conventional solid state reaction using high-purity SmAs, SmF_3, Fe, and Fe_2O_3 as starting materials. The raw materials were thoroughly ground and pressed into pellet. The pellet was wrapped in Ta foil, sealed in an evacuated quartz tube, and finally annealed at 1160 °C for 40 h.[11]

The positron lifetime spectra (PLS) were measured using a fast-slow coincidence system with a resolution of 210 ps. A 10 μCi ^{22}Na positron source was sandwiched between two pieces of identical samples. The samples were attached to the cold head of a closed-cycle He refrigerator and the temperature were controlled by a Temperature Controller (S331) with the error below 0.5 K. Each spectrum contains more than one million events.

The calculation of positron is an important component in the research of positron techniques. Compared with other methods, the SNA-FD method needs little time and has the same precision. It can solve complex vacancy type and become a broad calculation method in the international positron research.[12,13]

The Doppler-broadening annihilation 511 keV γ-rays of positron-electron pairing was detected using an HPGe Detector (ORTECGEM-10175) with energy resolution of 1.12 keV (full width at half maximum) at the 514 keV 85Sr γ-ray. Each spectrum was collected with a total count over 2×10^6 and characterized by the S parameter and W parameter which express the relative ratio of low momentum electrons and high momentum electrons annihilation with positrons, respectively. Higher S value for a given material suggests more annihilation with low-momentum electrons, while higher W suggests more annihilation with high-momentum electrons. Detailed descriptions of PALS and DBS can be found elsewhere.[14,15]

PLS experiments of SmFeAsO and $SmFeAsO_{0.82}F_{0.18}$ superconductor were first carried out at 298 k. Each spectrum was fitted by LIFETIME9[16] program and the variance in fit is less than 1.06. The results were summarized in Table I.

τ_1 is mainly associated with free positron annihilation in the bulk of the sample. According to the experiment results in Table I and two-state trapping model,[17]

$$\tau_1^{-1} = \tau_{\text{bulk}}^{-1} + (\tau_{\text{bulk}}^{-1} - \tau_{\text{trap}}^{-1})\frac{I_2}{I_1}, \tag{1}$$

TABLE I. Positron lifetime results.

Sample	τ_1 (ps)	I_1 (%)	τ_2 (ps)	I_2 (%)
SmFeAsO	151.6(2.2)	60.8(1.6)	290.3(7.3)	38.8(1.6)
$SmFeAsO_{0.82}F_{0.18}$	161.6(2.7)	74.5(1.7)	316.4(9.4)	25.1(1.8)

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$$\tau_2 = \tau_{trap}, \tag{2}$$

$$\tau_{bulk}^{-1} = I_1\tau_1^{-1} + I_2\tau_2^{-1}, \tag{3}$$

the bulk lifetime is evaluated for each sample, which are 187.0 and 185.1 ps for parent and superconductor, respectively. A long-lived component τ_2 was also obtained in the PLS, which must play an important role for superconductivity. Where is it from?

The bulk lifetime is usually obtained from a measurement on a crystal free from positron-trapping defects and is compared with a theoretical calculation to justify the goodness of the samples and the calculation employed. However, it is impossible in the present case because the high-T_c samples always contain the defects. Thus the bulk lifetimes were estimated using the Eq. (3) on a usual assumption of the two-state trapping model and compared with a standard theoretical model calculation which gives usually good agreement with experiments. Based on the SNA-FD method, positron bulk lifetimes and positron monovacancy lifetimes of perfect SmFeAsO and SmFeAsF crystals were calculated and the results are shown in Table II.

The results show that there is no observable change of the positron lifetime between SmFeAsO and SmFeAsF crystal, and the calculated bulk lifetime value of SmFeAsO and SmFeAsF crystal agrees well with the experimental value within the error tolerance. Furthermore, it indicates the long-lived component τ_2 may attribute to positron annihilation in the monovacancy of Sm with lifetime about 300 ps or in the complex-defect or in the region of grain boundary.

In order to obtain more information about which defects associated with τ_2, we carried out DBS measurements in SmFeAsO and $SmFeAsO_{0.82}F_{0.18}$ samples. The S parameter as a function of temperature in parent and superconductor sample is shown in Figs. 1 and 2, respectively. The error of the S parameter is all below 0.0007 which we obtained by continuous measuring one point for 16 times in 24 h. The S parameter of the parent sample is well fitted with two straight lines separated at about 150 K, the jump of which indicates that a tetragonal to orthorhombic phase transition[11,18] occurred at that temperature. Meanwhile, the unobvious change of the S parameter implies that the prominent arising of superconductor's S parameter with the increasing temperature is not only related to the tiny variation in lattice parameters. The S parameter of the superconductor sample shows two different slopes below and above superconductivity transition, which infers that the momentum distribution of electron annihilated with positron has varied.

TABLE II. The calculated positron lifetimes of SmFeAsO and SmFeAsF crystals.

Sample	Variety of vacancy	Lifetime (ps)
SmFeAsO	Bulk lifetime	173
	Fe monovacancy	210
	As monovacancy	242
	Sm monovacancy	280
SmFeAsF	Bulk lifetime	175
	Fe monovacancy	210
	As monovacancy	243
	Sm monovacancy	288

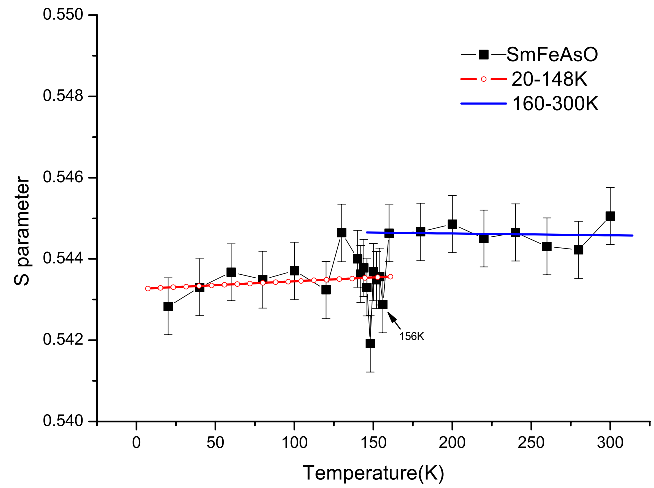

FIG. 1. (Color online) Temperature dependence of the S parameter in the parent sample.

The cross-point of two slopes is 68.5 K, much higher than T_c of 48.5 K, which may arise from the already existent Cooper pairs above T_c.[19]

It would also be interesting to find out how many defect types in the superconductivity transition. The number of different vacancy-type defects trapping positron in the material can be investigated though the linear relation between S and W.[20] If only a single type of vacancy is present, the W parameter depends linearly on the S parameter. From the dependence of parameters S and W shown in Fig. 3, it clearly exhibits that only one-type of defects appears in the superconductor sample through the superconducting transition. We can conclude that τ_2 may attribute to positron annihilation in the monovacancy of Sm with lifetime about 300 ps or in the complex-defect.

Based on the SNA-FD theory, we calculate the positron lifetime of SmFeAsO and SmFeAsF crystals with different types of vacancies. Combined with PALS and DBS measurements, we find out some results of $SmFeAsO_{1-x}F_x$ polycrystalline sample, summarized as follows:

1. The calculated positron bulk lifetime in polycrystalline $SmFeAsO_xF_{1-x}$ agrees well with the experimental results.
2. The temperature dependence of S-parameter shows a re-

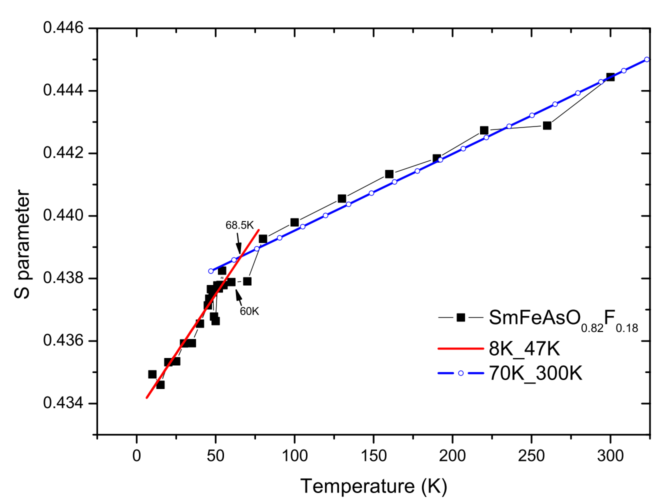

FIG. 2. (Color online) Temperature dependence of the S parameter in superconductor sample.

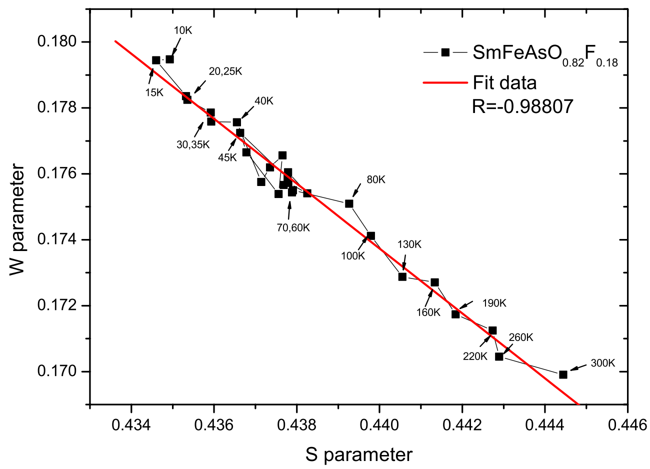

FIG. 3. (Color online) The dependence of S and W parameters.

markable difference of the parent and the superconductor samples.

3. At 68.5 K, much higher than T_c of 48.5 K, Cooper pairs might already exist.
4. Only one-type of defects exists in the superconductor sample through the superconductivity transition.
5. The long-lifetime component may attribute to positron annihilation in the Sm monovacancy or in the complex-defect.

But the accurate defects related to superconductivity still can't determined in the $SmFeAsO_{0.82}F_{0.18}$ sample, More experiments and better methods of data processing should be conducted in future. In any case, we are sure that this new class of materials will open a new avenue of research high-temperature superconductor.

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