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Study of Coincidence Doppler Broadening in Carbon Nanotubes*

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Abstract Coincidence Doppler broadening measurements of positron annihilation for multi-walled carbon nanotubes, double-walled carbon nanotubes, single-walled carbon nanotubes and graphite were performed. The ratio curves of the Doppler broadening for these samples to silicon were obtained. It is shown that there are distinct peaks at the position of $10\times10^{-3}m_0c$ for both carbon nanotubes and graphite, however the amplitudes of the peaks are not the same. We have the opinion that these peaks arise from the annihilation of positron with the $2s$ and $2p$ electron of carbon element.

Keywords: positron annihilation, coincidence Doppler broadening, carbon nanotubes

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1 Introduction

Carbon nanotubes (CNTs) can usually be viewed as hollow cylinders formed by rolling up graphite sheets, which are several nanometers in diameter and many microns in length. According to the sheet numbers of rolling, carbon nanotubes are classified as single-walled carbon nanotubes (SWNT), double-walled carbon nanotubes (DWNT) and multi-walled carbon nanotubes (MWNT) [1]. Carbon nanotubes aroused great interest in the research community because of their unique physical properties, such as the highest Young's modulus and tensile strength, the metallic or the semiconducting characteristics of a SWNT depending on the diameter and helicity [2,3].

The measurement of positron lifetime, the Doppler broadening and angular correlation of the annihilation radiation are three elemental techniques regarding positron annihilation, which are traditionally adopted to probe the electronic properties and geometric structures in solids [4]. The positron lifetime is inversely proportional to the density of the electron at the positron-electron pair annihilation site, while Doppler broadening and angular correlation of annihilation radiation reflect the momentum distribution of the electron annihilated with the positron. More exactly, the longitudinal momentum component of the electron (P_l) along the direction of the annihilated γ-photon emission is related to the Doppler broadening (ΔE) of the annihilation radiation energy by

$$P_l = 2\Delta E/c, \tag{1}$$

where c is the speed of light. In addition, many significant improvements can be achieved by coincidence Doppler broadening measurement using two HPGe detectors [5,6]. Since the peak-to-background ratio can be improved greatly, coincidence measurement leads to further investigations of the high momentum component of the Doppler broadening, corresponding to the annihilation of the core electron in the samples, which makes up the deficiency of the single Doppler broadening measurement.

Many studies on the properties and application of carbon nanotubes have been performed using various methods including positron annihilation spectroscopy. Positron lifetime measurements for carbon nanotubes were reported and most results show that CNT has a lifetime between 360 ps and 430 ps, owing to the annihilation of positron at the surface of the CNTs [7~9]. Additionally, ITO Y observed the difference of FWHM (Full Width at Half Maximum) of the electron momentum distribution between graphite, diamond, nanotubes, C60 and C70 fullerene by Doppler broadening measurements in 1999 [7], and the difference of the electron density distribution for the allotrope of carbon was indicated again with coincidence method in 2004, leading to a conclusion that the positron annihilates with different components of the π and σ electrons in these materials [10].

In this work, we have performed the coincidence Doppler broadening spectroscopy for SWNT, DWNT, MWNT and graphite at various temperatures between 120 K and 340 K, and presented that the positron annihilates with the $2s$ and $2p$ electron of carbon element based on the analysis of ratio spectrum of the measured samples to silicon.

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2 Experiment

All of the CNT samples with a purity of better than 95 percent were prepared as powder using CVD by Chengdu Organic Chemicals Co. Ltd. CAS. Before the measurements, each sample was pressed into a couple of disks with a diameter of 13 mm and a thickness of about 2 mm. The digital coincidence Doppler broadening (CDB) annihilation spectrometer, which uses two HPGe detectors with energy resolution of 1.12 keV (FWHM) at 514 keV ^{85}Sr γ-photon, was employed to measure the 511 keV γ-photon of positron-electron pairs as shown in Fig. 1. The advantage of this digital system is its better reliability and flexibility in off-line data analysis. Fig. 2 shows a two-dimensional Si CDB spectroscopy. Spectra for various CNTs and graphite were collected with total counts of 8×10^6. The coincidence Doppler broadening spectra were recorded for SWNT and MWNT at the temperature from 120 K up to 340 K with an interval of 20 K.

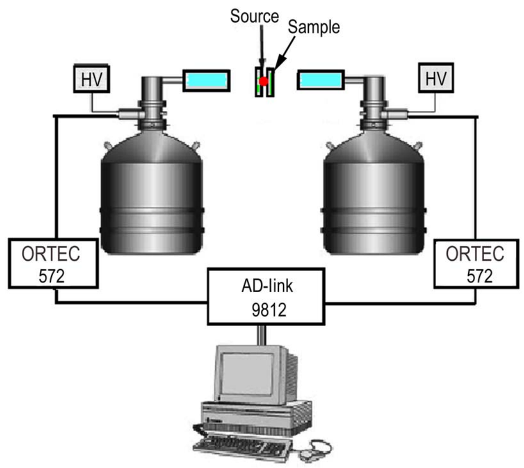

Fig.1 The set-up of the digital CDB spectrometer

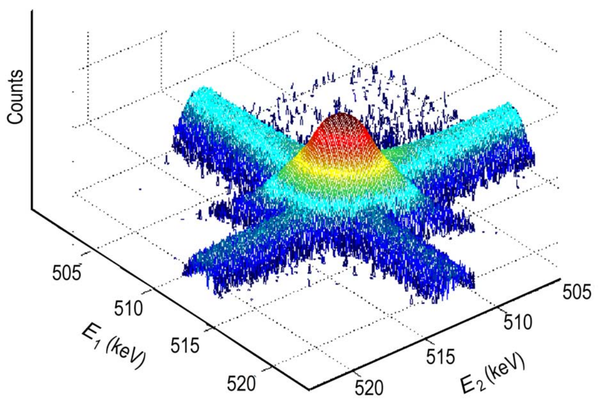

Fig.2 2-dimensional Si CDB spectroscopy (color online)

3 Results and discussion

Fig. 3 shows the ratio curves of the Doppler shift to silicon for SWNT in the temperature range between 120 K and 340 K. It is surprisingly found that the ratio curves for the various temperatures coincide well at the lower momentum region between zero and $15\times10^{-3}m_0c$, which corresponds generally to the valence electron annihilated with the positron. In addition, a peak is observed at the position of $10\times10^{-3}m_0c$, and the amplitudes of the peaks under various temperatures are the same. That is to say, variation of temperature does not change the peak position and the peak height, not having an effect on the electron structure of the SWNT. This phenomenon was also observed for the MWNT employed in our experiments, as shown in Fig. 4. On the base of this, the coincidence Doppler broadening spectra for DWNT and graphite at the room-temperature were performed and compared to the data of the SWNT and MWNT.

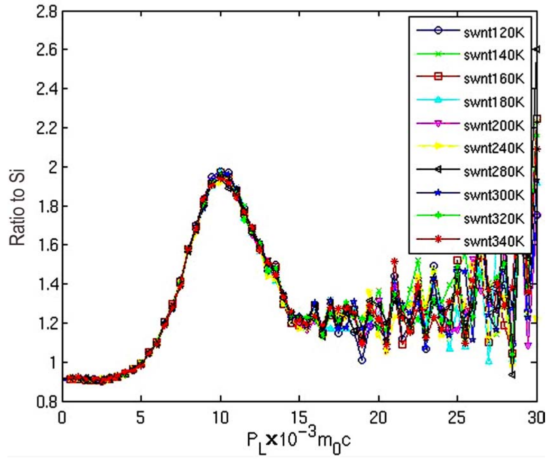

Fig.3 The ratio spectra of Doppler shift for SWNT to silicon at various temperatures (color online)

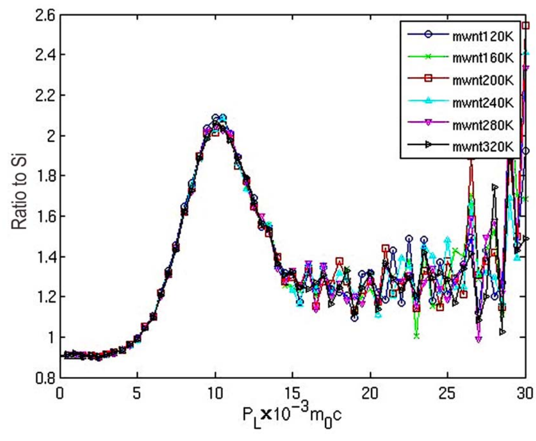

Fig.4 The ratio spectra of Doppler shift for MWNT to silicon at various temperatures (color online)

Fig. 5 shows the ratio curves of the Doppler shift to silicon for SWNT and MWNT at the temperature of 120 K, DWNT and for graphite at the room-temperature. It can be seen that all the samples have a peak at the point of $10\times10^{-3}m_0c$, but the amplitudes of various peaks for the samples are not the same.

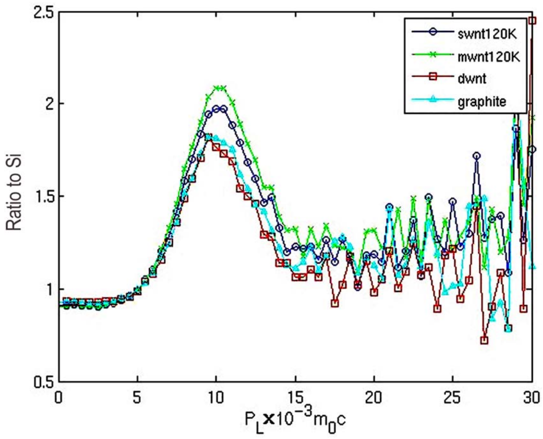

Fig.5 The ratio spectra of Doppler shift for CNTs and graphite to silicon (color online)

DENG W etc. have reported the ratio shape of the Doppler broadening for carbon, boron and beryllium second-period elements to silicon, describing that the Doppler peaks of three elements are positioned at the same point of $10\times10^{-3}m_0c$ and the amplitudes of the peaks is considerably different. However, the authors did not explain this phenomenon in detail at that time [11]. In 2009, DENG W etc. presented that the peak of the Doppler shift of graphite arises from the annihilation of the positron with the $2p$ electron of carbon element [12]. But we believe that the peaks of 10×10^{-3} m_0c for various CNTs and graphite come from the double contributions of both $2p$ and $2s$ electrons. Since the annihilation probability of the positron with the $2s$ and $2p$ electrons for CNTs and graphite is different, the height of the annihilation peak is not the same, reflecting the intrinsic difference between the electron structures of these samples. This opinion also explains the discrepancy of the peak height for carbon, boron and beryllium in DENG's work.

4 Conclusion

We have performed measurements of the Doppler broadening spectroscopy for SWNT, DWNT, MWNT and graphite by using coincidence method. The ratio curves for these samples to silicon were obtained. Changes of temperature do not affect the position and height of the specific peak in the CDB ratio curves for SWNT and MWNT. On the other hand, all the samples have the same position of Doppler broadening peak, although the height of the peak is different. We have the opinion that the discrepancy of the height of the peak between the measured samples arises from the different annihilation probability of the positron with the $2s$ and $2p$ electrons, reflecting the intrinsic difference between the electron structures of these samples.

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