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A High Resolution Investigation of the Anisotropic Electron-Positron Momentum Distribution in Diamond

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Abstract

We have performed a high resolution, high sensitivity study of the electron-positron momentum distribution in both natural and synthetic diamond, using the two-dimensional Doppler broadening technique of positron annihilation spectroscopy. The measurements were carried out with the <100>, <110> and <111> axes each separately aligned collinearly with the detectors so that the anisotropy of the momentum distribution could be mapped out. Small differences in the momentum profiles were amplified with subtraction spectra and compare well with results of a two-component density functional theory calculation of the electron-positron momentum distribution in the diamond lattice. There has been much debate regarding previously unexplained anomalies in the annihilation characteristics of the bulk positron configuration in diamond. These new results strongly support a model describing the bulk positron as delocalised and inhomogeneous, but with sufficient amplitude at the intra-bond region such that the annihilation characteristics are determined by the bond electron density and momentum distribution.

1. Introduction

Positron annihilation spectroscopy (PAS) has become a powerful tool for the study of bulk diamond, diamond surfaces and defects in the diamond lattice. This spectroscopy has, for example, been used to investigate isotopically pure synthetic diamond [1], electron-irradiated [2] and gamma-irradiated [3] diamond, diamond films [4,5] and large vacancy clusters in natural diamond [6]. The ability of the positron to probe optically active centres in natural and industrial grade diamond has been recently demonstrated [3,7] and much work has been carried out on the anisotropic electron-positron momentum distribution in diamond, both theoretically and experimentally [8,9,10].

The current paper reports on two-detector coincidence Doppler broadening measurements carried out on synthetic and natural diamond samples. The measurements were performed at different crystallographic orientations (<100>, <110> and <111>) to study the anisotropy of the electron-positron momentum distribution. The subtle anisotropies were enhanced with difference spectra. The results are compared directly to the predictions of a two-component density-functional theory (DFT) calculation of the electron-positron momentum distribution for the case of the delocalised positron in the perfect diamond lattice. The many-body aspects of the electron-positron correlation were treated in the independent particle model (IPM). The theoretical calculation has been presented in reference [11] and the measurements described below are discussed in greater detail in references [12,13].

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The main advantage of the two-detector technique introduced by Lynn *et al.* [14] over the conventional one-detector technique is that peak to background ratios of better than 10^5:1 are possible. Thus high momentum components can be efficiently observed in the energy spectra. In addition, the energy resolution of the spectrometer can be improved by a factor $\sqrt{2}$ and the binding energy of the electron-positron system to the host can be measured. As a result of the increased resolution and sensitivity of the technique, the experiments represent a dramatic enhancement over the previous one-detector measurements (ref. [9]) of the anisotropic electron-positron momentum distribution in diamond. The reader is referred to [15,16] for further discussion regarding the technique.

2. Results

The measurements were carried out with the <100>, <110> and <111> axes each separately aligned collinearly with the detectors so that the anisotropy of the momentum distribution could be mapped out. Each two-dimensional spectrum contained approximately 6×10^6 counts and took 20 hours to collect. The electron-positron momentum profiles were extracted from the spectra and normalised to unity before difference spectra were taken. The spectra were then folded about 511 keV for improved statistics and clarity. The difference spectra taken for the synthetic diamond are presented in fig. 1. The results for the natural diamond are identical.

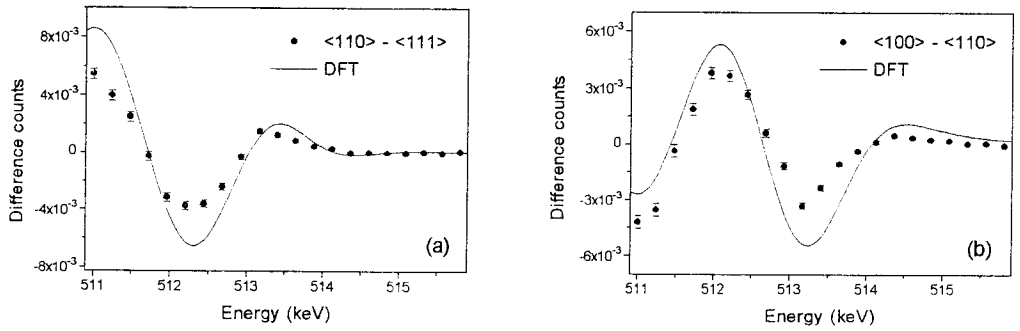

Figure 1. Difference spectra taken for the three main crystallographic orientations in diamond. Subtle anisotropies in the electron-positron momentum distrubtion are clearly observed. (a) <110> - <111> (b) <100> - <110> . The predictions of the DFT calculation have been included for direct comparison.

Fig. 1(a) indicates that the momentum distribution projected on to the <111> axis is broader in the middle regions (from roughly 511.5 keV to 513 keV) but without the very high momentum component tails of the <110> axis above 513 keV. This agrees well with the one-detector measurement reported earlier [9] where minima in the S-parameter were observed for projections of the momentum distribution on to the <111> axes. The data presented in fig. 1(b) is the clearest example of the advantages a two-detector coincidence measurement has over a one-detector measurement. In the S-parameter analysis of the data taken in the one-detector measurement, the projections of the momentum distribution onto the <100> and <110> axes yielded almost identical S-parameters. However, the two-detector investigation presented above reveals clear differences in the momentum profiles. The presence of two minima at approximately 511 keV and 513.2 keV in this difference spectrum reflects the anisotropic nature of the electron-positron momentum distribution in the diamond lattice.

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Included in fig. 1 are the corresponding difference spectra obtained from the DFT calculation of the electron-positron momentum distribution in diamond and the qualitative agreement is excellent. Quantitatively, however, the curves differ significantly. After convoluting the theoretical curves with a Gaussian model of the energy resolution of the spectrometer, they show a variation of approximately 1.5 times greater than the experimental curves. This discrepancy is attributed partly to the fact that the experiment was performed at room temperature whereas temperature effects were ignored in the theoretical calculation. Temperature is expected to broaden the momentum profiles and thus decrease the magnitude of any variations. Dannefaer *et al.* [17] recently demonstrated this effect in Doppler broadening studies of several diamond-like semiconductors. The presence of defects in the diamond lattice also contributes significantly to the quantitative discrepancy. Subsequent lifetime measurements on the sample revealed a 14(1)% trapped positron intensity. The trapped positron contribution to the electron-positron momentum distribution cannot be reliably extracted and so will significantly reduce any observed anisotropy. In addition, the (approximately) 4^0 angular resolution of the spectrometer, the IPM treatment of the electron-positron correlation and the use of pseudopotentials to calculate the electron density contribute.

Fig. 2 shows the DFT calculation of the electron-positron density overlap distribution in the [110] plane.

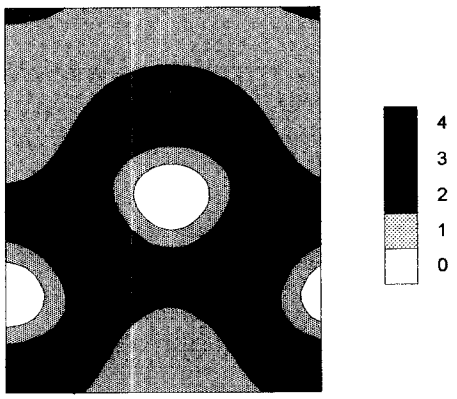

Figure 2. The overlap distribution of the electron-positron densities in a [110] plane showing the pile-up in the bond region. The contour levels scale from 0 (white) to 4 (black) in arbitrary units. The dashed line represents the bond-region / interstitial region boundary chosen for the annihilation site estimate discussed in the text.

This distribution reveals a maximum in the bond regions. To compare the total annihilation intensity in the bond and interstitial regions, the bond region was defined as any site with an electron density parameter $r_s < 1.34$ (atomic units). The dashed line in fig. 2 indicates the chosen interface between these two regions. The total probability weights were then calculated. This rough method of calculation yielded a bond region annihilation to interstitial region annihilation ratio of 1:1 (± 5%). Thus despite repulsion due to the carbon cores there is a high probability of annihilation in the bond region. It is suggested that this explains the broad momentum distribution observed for the short lived positron configuration in diamond.

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3. Conclusions

The two-detector coincidence Doppler broadening technique of positron annihilation spectroscopy allows a high resolution measurement of the electron-positron momentum distribution in condensed matter. Measurements have been performed on a synthetic diamond sample as a function of crystallographic orientation and the anisotropy of the electron-positron momentum distribution was investigated. The experimental results are in good agreement with a density-functional theory calculation of the electron-positron momentum density distribution.

It has been demonstrated that the broad momentum distribution of the bulk positron configuration in the diamond lattice can be attributed to positron annihilation with electrons in the carbon-carbon bond region. These new results indicate that although the positron density is small in the bond region due to core repulsion, the extreme localisation of the electron density there compensates for this such that the annihilation characteristics of the positron in bulk diamond are determined by the bond electrons. This model can therefore explain simultaneously the high initial population, short lifetime and broad momentum distribution of the bulk positron state. In pure diamond, this should be the only state for deeply implanted positrons. The spread in the reported bulk positron lifetimes, the appearance of other lifetime components and the observation of positronium formation in the diamond lattice are due to defect related positron configurations.

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