MODELING POSITRON–ELECTRON CORRELATIONS IN SOLIDS

Lise GILGIEN¹, Roberto CAR¹ and David M. CEPERLEY²

¹ IRRMA, PPH-Ecublens, CH-1015 Lausanne, Switzerland
² NCSA, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

Keywords: positron–electron correlations, defects in semiconductors, Quantum Monte Carlo, first-principles molecular dynamics and 2D-ACAR.

ABSTRACT

Two new LDA parametrizations for positron–electron correlations are proposed on the basis of Quantum Monte Carlo results in an homogeneous 2-component jellium. They are tested towards Quantum Monte Carlo results in a simple inhomogeneous model before being applied to the study of positron trapping at As vacancies in GaAs. In both systems, one parametrization overestimates the positron localization whereas the other one underestimates it.

INTRODUCTION

The study of positron properties in condensed matter systems, particularly of the possibility for positron states in semiconductors to be either extended in the bulk or localized at defect sites, requires a good understanding of positron–electron correlations. The standard approach for dealing with positrons in inhomogeneous systems is a 2-component Density Functional Theory (DFT) in the Local Density Approximation (LDA). Existing formulations based on this approach have several problems, some of which are related to the imperfect knowledge of positron–electron correlations, even in homogeneous systems.

This work aims at improving the theoretical approaches to deal with positrons and electrons in solids. In particular, the main objective is to find a better LDA parametrization for positron–electron correlations on the basis of accurate results in homogeneous systems. To achieve these goals, positron–electron correlation effects in an homogeneous 2-component jellium have been studied within Quantum Monte Carlo (QMC) techniques. New LDA parametrizations have been constructed on the basis of these results. They have been tested in a simple inhomogeneous system towards more accurate Quantum Monte Carlo results. They have finally been applied to the study of positron trapping at As vacancies in GaAs.

LDA PARAMETRIZATIONS

Quantum Monte Carlo calculations in the homogeneous 2-component jellium have been performed for an extended range of densities and positron concentrations. In spite of the fact that these results constitute probably the largest data base for positron–electron correlation energies, it is not sufficient to uniquely define an LDA parametrization for positron–electron correlation. Two different parametrizations have therefore been proposed. They will be refered to as 2CP1 and 2CP2 and are represented in Fig. 1.
Figure 1: Positron–electron correlation energies per particle as a function of $|r_s| = \sqrt{(r_s^+)^2 + (r_s^-)^2}$, for different values of $\xi$ where $\xi = r_s^-/r_s^+$ if $r_s^- \leq r_s^+$ and $r_s^+/r_s^-$ otherwise. DMC results obtained with 2-component plasma type trial wavefunctions are represented by dots, whereas the 2CP1 (upper pannel) and the 2CP2 (lower pannel) parametrizations are represented by solid lines.
SIMPLE INHOMOGENEOUS SYSTEM

The simple inhomogeneous system considered for testing the LDA parametrizations towards accurate Quantum Monte Carlo results consists of a spherical vacancy in a jellium. Fig. 2 shows the positron localization as a function of the vacancy radius, as obtained by Quantum Monte Carlo techniques and DFT-LDA calculations using both the 2CP1 and the 2CP2 parametrizations. It can be seen that 2CP1 slightly overestimates the positron localization whereas 2CP2 underestimates it.

![Graph showing positron localization as a function of vacancy radius](image)

Figure 2: Number of positrons (upper panel) and electrons (lower panel) contained in a sphere of radius 1 a.u. centered on the vacancy center, for \( r_v^- = 3 \) a.u. and as a function of the vacancy radius. Quantum Monte Carlo results are represented by solid lines, whereas DFT-LDA results are represented by dotted lines. DMC stands for Diffusion Monte Carlo and PIMC for Path Integral Monte Carlo (\( T = 0.0125 \) Ry for \( r_v \leq 1.5 \) a.u. and \( T = 0.025 \) Ry for \( r_v \geq 1.5 \) a.u.)

As VACANCIES IN GaAs

It can be concluded from the DFT-LDA results obtained in GaAs that an extended positron state leads to a 2D-ACAR spectrum with a bone like feature in the center, whereas a localized positron state results in a more spherical 2D-ACAR spectrum. Fig. 3 therefore shows that experimentally, the positron state is delocalized in the bulk of GaAs, whereas it is localized at both the neutral and the singly negative As vacancies. However, 2CP1 leads to localized positron states (even in the bulk), whereas 2CP2 leads to delocalized positron states (even in \( V^-_{As} \)). This confirms that 2CP1 (resp. 2CP2) overestimates (resp. underestimates) the positron localization. This results in an overestimation (resp. underestimation) of the differences in
Table 1: Experimental [2] and theoretical positron lifetimes obtained for the bulk of GaAs, $V^-_{As}$ and $V^0_{As}$ (in ps). All lifetime computations used the AP paratermization for the contact density and included the core electron charge density. The inaccuracies induced by the choice of the fit for the core charge density are of the order of ±4 ps.

positron lifetimes between the bulk and the vacancies with 2CP1 (resp. 2CP2), as can be seen from Table 1.

CONCLUSION

Since both 2CP1 and 2CP2 fit the Monte Carlo results in the homogeneous systems equally well (see Fig. 1), the results obtained in vacancies in jellium and in GaAs indicate that it will probably be possible to construct an LDA parametrization capable of predicting whether a positron is localized or not, once additional Monte Carlo results will be available for the homogeneous system. Work is in progress with this aim in view.

ACKNOWLEDGEMENTS

References


Figure 3: Experimental [1] and theoretical 2D-ACAR spectra integrated along the [110] direction obtained for the bulk (left column), $V_{\text{As}}$ (middle column) and $V_{\text{As}}^0$ (right column). The experimental results are displayed in the first row, theoretical results obtained with the 2CP1 and 2CP2 parametrizations in the second and third row, respectively. The momenta $p_i$ are related to the measured angles $\theta_i$ by $p_i = mc\sin \theta_i \simeq mc\theta_i$. All theoretical spectra were computed using the AP parametrization for the contact density. They do not include any core electron effects.