



Insight into the electron–positron correlations in metals through the looking glass



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ABSTRACT

A semi-empirical analysis of the positron annihilation experimental spectra indicates for a strong sensitivity of the two-particle electron–positron (e – p) enhancement factor to the $l=s, p, d, f$ character of the initial electronic state [1,2]. The essential discrepancy between the models consists in the dependence of the relevant correlation functions on the energy of the annihilating electron. The present contribution contains a theoretical study of the e – p enhancement factors for s, p, d and f states as a function of the electron energy. The slope of the resulting characteristics is directly related to the degree of localisation of the s, p, d and f electrons in the electron density of states. This effect occurs especially for d electrons in transition metals, in favour to the approach of Ref. [1].

The energy dependence of the two-particle correlation functions is also a source of controversy between various theoretical approaches. The energy dependent enhancement factors describe properly the positron interaction with delocalised s and p electrons, but this approach overestimates the high momentum components of the e – p momentum densities, dominated by the localised d and f states. On the contrary, the calculations that employ the energy averaged enhancement factors match better with experiment for localised d and f electrons, but they hardly reproduce experimental spectra for nearly-free electron populations. An attempt to visit two sides of the looking glass is made in the theory of the present work. The model combines the properties of both approaches. The resulting e – p momentum densities and enhancement factors are in good agreement with the experimental data for simple, noble and transition metals, both in the low and high momentum region.

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1. Introduction

The positron annihilation spectroscopy is a sensitive and effective technique to probe the electronic structure of solids. In particular, the positron lifetime is a unique characteristic of the electron charge density, $n(\mathbf{r})$, in the Wigner–Seitz cell and the angular correlation of the positron annihilation radiation (ACPAR) or the coincidence Doppler broadening experimental data carry a useful information on the electron momentum density (EMD) in the host material, $\rho^{EMD}(\mathbf{p})$ [3,4]. This information is, however, modified by two effects: the strong electron–positron attraction and the deviation of the positron wave function, $\psi_+(\mathbf{r})$, from the single plane wave (with momentum $\mathbf{k}_+=\mathbf{0}$). Fortunately, the presence of the positron hardly shifts the positions of the Fermi momenta, \mathbf{p}_F [5], that evidence as a Fermi surface (FS) breaks in the $\rho^{EMD}(\mathbf{p})$ and e – p momentum density (MD), $\rho^{e-p}(\mathbf{p})$. In consequence, the shape of the FS, as measured by the positron annihilation spectroscopy, remains unchanged with respect to the one

retrieved from the EMD.

The interpretation of the positron annihilation experimental data relies on the principle of the energy and momentum conservation of the annihilating pair. Therefore, the usefulness of the information, recovered from the comparison of the experimental and theoretical positron annihilation characteristics, is crucially dependent on the accuracy of the approximation incorporated to the calculation of (1) the electronic structure in the host material, (2) the positron distribution and (3) the enhancement of the density of individual electronic states on the positron site. The sensitivity of the e – p MD to the details of the electron and positron band structure method and to the positron distribution has been pointed out by Majumdar [5] and intensively studied for contributions from both the core and valence electrons (*cf. e.g.* [2,6–10] and references cited therein). The important point in the study of the EMD and e – p MD in solids is the choice between the extended and reduced zone scheme [2,7]. It is well known, that for any fixed crystallographic direction there are some valence bands (well defined by the symmetry rules [11]) which give no contribution to the EMD and the relevant e – p MD in the extended

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zone scheme for momenta along this direction. In contrast to the extended zone scheme, the spectra, folded to the first Brillouin zone within the Lock–Crisp–West (LCW) method [12], contain the contributions from each of the occupied valence bands. In consequence, all the FS breaks well evidence in the LCW folded spectra for momenta along each of crystallographic directions, even for these bands which give no contribution along a chosen direction to the EMD and e - p MD in the extended zone scheme. Furthermore, in the present work a direct relation between the l -projected electron density of states (DOS) and the l -character contribution to the LCW folded EMD is derived. This property is generalized for the electron–positron system and it is shown that the relevant e - p enhancement factors (EF) reflect the degree of localisation of l states in the electron DOS. On the other hand, the high momentum components of the e - p MD are cumulated in the LCW folded spectra. Therefore, the information on the e - p enhancement factors in the individual Brillouin zones is partially lost in the LCW folded spectra. For this reason an application of both the reduced and extended zone scheme is highly required in a study of the e - p MDs and EFs.

The effect of the e - p interaction, leading to the formation of an electron screening cloud around the positron, is the third of ingredients that are essential for the interpretation of the positron annihilation data in terms of the EMD. Due to a strong Coulomb attraction between the (positively charged) positron and surrounding electrons, the initial density of any individual electronic state i in the host material, $|\psi_i(\mathbf{r})|^2$, is strongly enhanced at the positron position, \mathbf{r} , by a factor $\gamma_i(\mathbf{r})$. Throughout the present work, the factor $\gamma_i(\mathbf{r})$ is called the *partial correlation function*. In general, this quantity depends on both the initial electron state i and the positron position, \mathbf{r} . It should be noted here that the functions $\gamma_i(\mathbf{r})$ reflect not only the initial energy and momentum of the individual electronic states, i , as well as the electron density distribution in the host material, $n(\mathbf{r})$. Owing to the fact that the nearly-free sp -like electrons exhibit different screening abilities than the localised d or f states, the pair correlation functions depend also on the s,p,d and f character of the annihilating electron [1,2,10]. The change in the total electron charge density on the positron position, $n(\mathbf{r})$, is described by the *total correlation function*, $g(\mathbf{r})$. It is obvious that the function $g(\mathbf{r})$ is a weighted average of $\gamma_i(\mathbf{r})$, where the functions, $|\psi_i(\mathbf{r})|^2/n(\mathbf{r})$ are the weights.

The central goal of the present work is to study the partial and total correlation functions in crystalline solids, especially for the localised *versus* delocalised electron populations. Calculation of the functions $\gamma_i(\mathbf{r})$ and $g(\mathbf{r})$ is, however, a complicated many-body problem, which has not been solved exactly even for such a simple system as the homogeneous electron gas of the density $n_0 = p_F^3/(3\pi^2)$, in which the electron wave functions $\psi_i(\mathbf{r})$ are the single plane waves, associated with momentum \mathbf{p} and energy p^2 . Moreover, various theories (for review see e.g. [13–17] and references cited therein) lead to different results for the partial correlation functions in an electron gas, $\gamma^h(p^2, n_0)$, and to alternative parameterisations of the relevant total correlation functions, $g^h(n_0)$. In general, the function $\gamma^h(p^2, n_0)$ increases as a function of the initial electron energy, p^2 , according to the well-known Kahana's formula [13],

$$\gamma^h(p^2, n_0) = a(n_0) + b(n_0)(p^2/p_F^2) + c(n_0)(p^2/p_F^2)^2.$$

Except for the momenta in the close vicinity of the Fermi momentum p_F [14,16], this formula is well satisfied both in an electron gas as well as in the nearly-free electron systems (e.g. alkalis). Once again, the total correlation function in jellium, $g^h(n_0)$, is a weighted average of its partial counterparts.

The e - p correlation functions in crystalline solids are modified additionally by the periodic potential of the lattice, as compared to

their counterparts in an electron gas. This effect has been taken into account within the e - p density functional theory [15] in the majority of calculations of the positron annihilation characteristics in real systems as well as in the theory of the present work. More precisely, the partial or total correlation functions have been approximated by their counterparts in an electron gas of density $n(\mathbf{r})$, in a local or non-local way (see e.g. Refs. [6,8–10,17–22] and references cited therein). In general, the pair correlation functions in solids have been modelled within various theories as the functions of either the initial electron energy, E_i , or the Bloch state (see e.g. Refs. [1,6,8,17–21,23]), or the l -character of this state [1,2,10,21–23], or the lattice effects [20,24] as well as of the electron density distribution in the Wigner–Seitz cell, $n(\mathbf{r})$ [6–10,17–23] (and references cited therein).

In principle, one can distinguish between two types of approximations: those which base on the state-selective partial correlation functions, $\gamma_i(\mathbf{r})$, against those which utilise the i -independent quantities, like the total correlation functions, $g(\mathbf{r})$, or $\gamma_0(\mathbf{r})$ (which refers to electrons at the bottom of the conduction band), successfully employed for core electrons [6,10,19]. The essential question is which of these approaches provides better description of the e - p interaction in real systems. The answer follows from the comparison of the resulting e - p MD, $\rho^{e-p}(\mathbf{p})$, with its counterpart reconstructed from the experimental ACPAR data. Here a direct account for the e - p correlation effect is given by the momentum dependent EF, $\varepsilon(\mathbf{p})$. This quantity is usually defined as a ratio of $\rho^{e-p}(\mathbf{p})$ to its counterpart obtained within the independent particles model (IPM), $\rho^{IPM}(\mathbf{p})$. More specifically, $\varepsilon(\mathbf{p}) = \rho^{e-p}(\mathbf{p})/\rho^{IPM}(\mathbf{p})$.

In the sp -like metals (e.g. alkalis, Al, Mg) as well as in Si, Cu, Zn or Ag, the e - p EFs, $\varepsilon(\mathbf{p})$, extracted from the experimental ACPAR data [2,4,18,23,25–28], exhibit the Kahana-like increasing slope for momenta inside the central FS. This strong momentum dependence of $\varepsilon(\mathbf{p})$ transfers to the dependence of the two-particle partial correlation functions on the electron energy, E_i . The relevant parameters, $\gamma_i(\mathbf{r}) = \gamma(E_i, \mathbf{r})$, have been intensively studied in Refs. [1,10,18,21–23]. It should be stressed here that it is impossible to obtain such an increasing slope of the e - p MD and EF within any theory based on $g(\mathbf{r})$ or $\gamma_0(\mathbf{r})$, determined either within the local density approximation (LDA) or non-locally [9,17]. The experimental ACPAR data [1,2,23,25–28] and theoretical study [8,10,18–21] indicate for a vital importance of a direct energy-dependence of the two-particle correlation functions in the nearly-free electron systems.

When the d (f) character increases in the electron DOS near the FS, the slope of the e - p EFs inside the central FS becomes less momentum dependent and the experimental e - p MDs are more and more IPM-like [1,2,9,17–24,29–32]. Approximation of the two-particle correlation functions, $\gamma_i(\mathbf{r})$, by the parameters less dependent on the electron energy, E_i , than $\gamma(E_i, \mathbf{r})$ (e.g. by $g(\mathbf{r})$ [9,17,20–22]) improves agreement between theory and experiment in the low momentum region. Also an implementation of the lattice effects into the original Kahana equation [24,32] essentially suppresses the momentum dependence of the e - p EFs for localised electron populations. This effect has been for the first time discussed by Fujiwara et al. [24] for the case of a simple two-band model.

The idea of the energy and l character dependent correlation functions originates from the early papers by M. Šob [23], who adapted the Kahana-like correlation function in an electron gas [13] to the case of the e - p interaction in a crystal lattice. Author assumed the energy dependence of the e - p enhancement factors in a Kahana-like form in which the kinetic energy of the annihilating pair, p^2 , has been replaced by the initial energy of the annihilating electron, E . This concept was independently published in the work by Mijnders and Singru and developed few years

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