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## LETTER TO THE EDITOR

# A local-density study of the electron–positron interaction in transition metals by positron annihilation

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**Abstract.** A theoretical local-density approach for treating electron–positron interaction in transition metals is presented and the enhancement effect for d bands in Cr, Ni, Cu and Pd is discussed. The approach presented is physically transparent and efficient—it explains the de-enhancement of d-bands observed earlier by Singh and co-workers, but gives less credit to the corresponding interpretation carried out recently by Jarlborg and Singh.

An important problem in interpreting the results of angular correlation of positron annihilation radiation concerns the influence of the electron–positron interaction on the two-photon momentum density distribution (TPMD),  $\rho(\mathbf{p})$ .

The first useful theory of  $e^-e^+$  interaction in an electron gas was proposed by Kahana [1]. His results may be written in terms of momentum-dependent enhancement factor  $\varepsilon(\mathbf{p}) = \rho(\mathbf{p})/\rho_{\text{IPM}}(\mathbf{p})$  given by

$$\varepsilon(\mathbf{p}) = a + b\rho^2/\rho_{\text{F}}^2 + c\rho^4/\rho_{\text{F}}^4 \quad (1)$$

where  $\rho_{\text{F}}$  is the Fermi momentum,  $a$ ,  $b$  and  $c$  are constants dependent on the electron density in a given material, and the momentum density in the independent particle model approximation (IPM) is expressed as

$$\rho_{\text{IPM}}(\mathbf{p}) = \sum_{k,n}^{\text{occ}} \left| \int \psi_{k,n}(\mathbf{r}) \psi_+(\mathbf{r}) e^{-i\mathbf{p}\cdot\mathbf{r}} d\mathbf{r} \right|^2 \quad (2)$$

with  $\psi_{k,n}(\mathbf{r})$  denoting the wavefunction of an electron with wavevector  $\mathbf{k}$  in the  $n$ th band and  $\psi_+(\mathbf{r})$  representing the wavefunction of a positron in the ground state.

In transition metals, where the localised d-like electrons are present, the simple expression (1) for the enhancement factor is no longer valid and a more general theory should be developed.

Nevertheless one can try to generalise this equation and extend it to more complicated electronic structures. For example Šob [2] and Mijnaerends and Singru [3] independently proposed an energy-dependent enhancement factor of the form

$$\varepsilon(E_{k,n}) = a + bE_{k,n}/E_{\text{F}} + cE_{k,n}^2/E_{\text{F}}^2 \quad (3)$$

where, in comparison with equation (1), the ratio  $p^2/p_{\text{F}}^2$  is substituted by the ratio of the electron energy  $E_{k,n}$  and the Fermi energy  $E_{\text{F}}$ , both counted from the bottom of the conduction band.

A substantial generalisation of equations (1) and (2) has been proposed by Daniuk and co-workers [4, 5]. In these papers the ratio  $p^2/p_F^2$  in the electron gas is substituted by the ratio of local kinetic energies  $X_{k,n}(\mathbf{r}) = [E_{k,n} - V(\mathbf{r})]/[E_F - V(\mathbf{r})]$ , where  $V(\mathbf{r})$  is the crystal potential and the enhancement factor is applied locally in the calculation of the overlap integral between electron and positron wavefunctions according to the formula

$$\rho(\mathbf{p}) = \sum_{k,n}^{\text{occ}} \left| \int \{\varepsilon[r_s(\mathbf{r}), X_{k,n}(\mathbf{r})]\}^{1/2} \psi_{k,n}(\mathbf{r}) \psi_+(\mathbf{r}) e^{-i\mathbf{p}\cdot\mathbf{r}} d\mathbf{r} \right|^2 \quad (4)$$

with  $r_s(\mathbf{r})$  being the local electron density parameter.

In order to perform the integration in equation (4) a complete knowledge of the enhancement factors is needed for all electron densities occurring in a real metal, and also inside the ionic core. Such calculations of  $\varepsilon$  have been performed for  $0.1 \leq r_s \leq 2$  by Rubaszek and co-workers [6]. Equation (4) applied to Zn gave a good agreement between theoretical and experimental positron-annihilation spectra [4, 5].

The investigations presented in this paper were motivated by very interesting results of Singh and co-workers [7] and Jarlborg and Singh [8] concerning 2D angular correlation experiments in Ni. The authors of the former study tried to interpret their results of combining IPM with energy-dependent enhancement factors. Surprisingly, in order to get a satisfactory agreement between experiment and theory, they had to accept a negative energy characteristic of the enhancement function of the d-states, leading even to the de-enhancement of the annihilation rate of the d states near  $E_F$ .

In the present Letter, we discuss the enhancement of d electrons in Cr, Ni, Cu and Pd. As our treatment is more general than that in [8], we are able to explain the increase of the d-electron enhancement factor throughout the d band in Cr, which was recently observed experimentally [9], as well as the negative energy characteristics of the enhancement factor (i.e. the relative de-enhancement of the annihilation rate) of electrons at the top of the d band in Ni and Pd reported in [7, 8].

Further, we show that the theory [8] always gives a d-electron enhancement factor decreasing with energy, which seems to be an obvious consequence of the behaviour of electron wavefunctions within the d band.

For non-hybridised d bands the enhanced TPMD is proportional to

$$L_2^2(E_{k,n}, p) = \left| \int_0^S \{\varepsilon[r_s(\mathbf{r}), X_{k,n}(\mathbf{r})]\}^{1/2} R_2(E_{k,n}, r) R_+(r) j_2(pr) r^2 dr \right|^2 \quad (5)$$

where  $R_2(E_{k,n}, r)$  is the radial wavefunction of the d electron,  $R_+(r)$  corresponds to the positron radial wavefunction,  $j_2(pr)$  is the Bessel function and  $S$  represents the Wigner-Seitz radius. Electron wavefunctions were computed on the basis of published potentials [10] and positron wavefunctions were obtained from the same potentials but without exchange and with opposite sign. That is why the ratio  $B_2(E_{k,n}, p) = [L_2(E_{k,n}, p)/L_2^{\text{IPM}}(E_{k,n}, p)]^2$  represents an effective enhancement factor in this case. The d bands which contribute to the TPMD by the largest amount (e.g.  $\Delta_1$ ,  $\Sigma_1$ ,  $\Lambda_1$ ) hybridise strongly with the s band and, therefore, the TPMD for d bands as such cannot be defined in a real d metal. In the case of hybridised bands, the overall enhancement is dependent on the relative weights of different  $L_1(E_{k,n}, p)$  in the annihilation amplitude (see e.g. [11]).

Nevertheless, the investigation of the 'partial' enhancement factor  $B_2(E_{k,n}, p)$  itself brings us a deeper insight into the  $e^-e^+$  interaction in d metals.

In the present calculations we use the values of  $\epsilon$  obtained in [6], where the  $X(r)$  is employed instead of  $(p/p_F)^2$ .

The results for the function  $B_2(E_{k,n})$  are presented in figure 1. In all the metals considered, except Cr, we can observe a weaker or stronger relative de-enhancement effect at the top of the d band (in the case of Ni and Pd this is in agreement with the experimental results of [7, 8]).

Explanation of the above de-enhancement effect is clearest when we assume that  $\epsilon$  depends on  $r_s(r)$  only (i.e.  $X(r) = 0$  in equation (5)). Such simplification of our approach was considered in [8] to explain the relative de-enhancement effect in Ni.

Figure 2 shows the radial variation of  $\{\epsilon[r_s(r), 0]\}^{1/2}$  and the radial wavefunctions of the positron and d electrons for the bottom, the centre and the top of d band in Cr, Ni, Cu and Pd. It can be seen that the enhancement factor has the same effect as a further localisation of the positron wavefunction near  $S$ . At the bottom of the d band the electron wavefunction is relatively delocalised and there is a large overlap with  $R_+$  and  $\epsilon^{1/2}$ . At the top of the d band the electron wavefunction is more localised and there is a smaller effect due to the enhancement. As a result the total integrated enhancement of the d band will be stronger for more delocalised electrons at the bottom of the d band than at the top of the d band.

However, it is known that the Wigner–Seitz rule [13] states that an l band can be formed in the energy range where the radial logarithmic derivative at the Wigner–Seitz sphere

$$D_l(E) = [S/R_l(E, S)]\partial R_l(E, r)/\partial r|_{r=S} \quad (6)$$

is negative. Usually the bottom ( $E_b$ ), the centre ( $E_c$ ) and the top ( $E_t$ ) of an l band are just defined as follows:

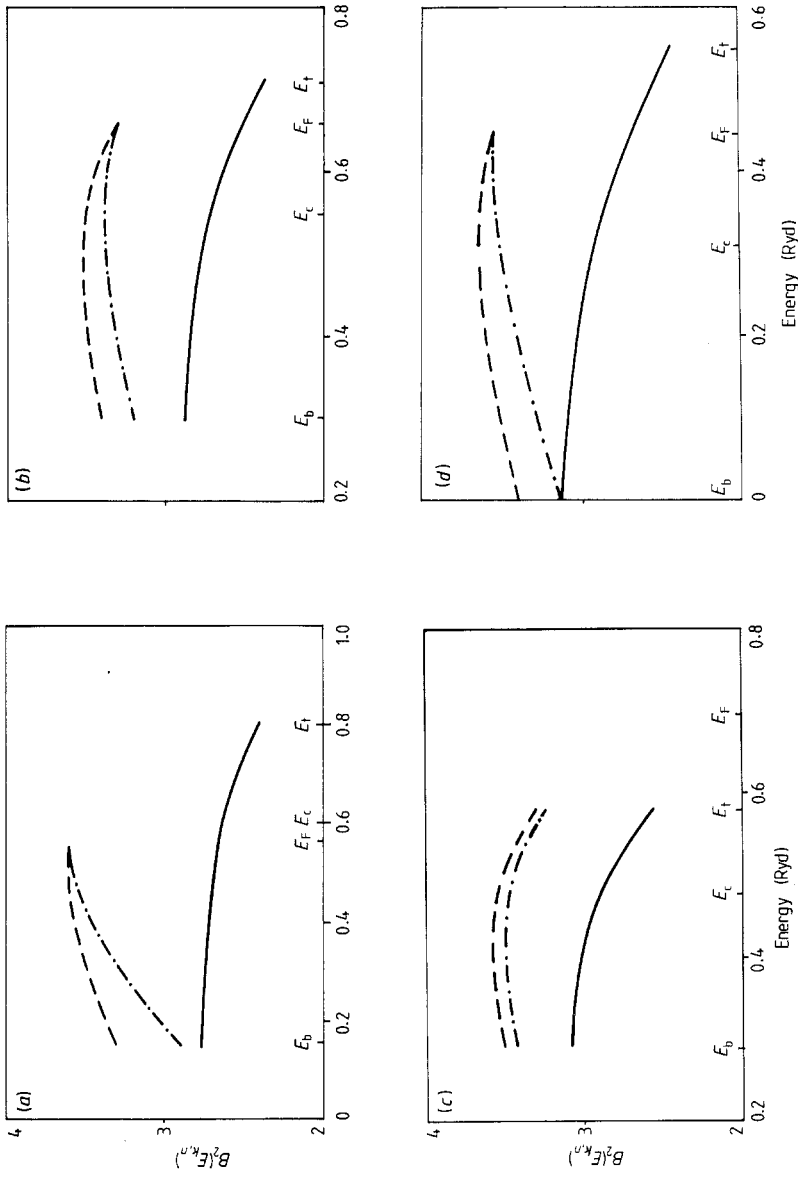
$$D_l(E_b) = 0 \quad D_l(E_c) = -l - 1 \quad D_l(E_t) = -\infty. \quad (7)$$

Therefore the results obtained for Cr, Ni, Cu and Pd (figure 2) as concerns the behaviour of wavefunctions of the electrons near  $S$  are rather general and consequently because  $\{\epsilon[r_s(r), 0]\}^{1/2}$  is an increasing function of  $r$  [14], the explanation of the relative de-enhancement effect given in [8] seems to be general as well but quite obvious, i.e. it concerns all metals with d bands or even any band at all. From experiment, however, we know that usually (especially for s bands) an enhancement effect (instead of the de-enhancement) is observable [2–6, 15, 16]. That is why an assumption that the enhancement factor depends only on  $r_s(r)$  seems to be too crude. Such treatment always gives a strong relative de-enhancement effect which is weakened when the energy dependence in  $\epsilon$  is taken into account (see figure 1), and as follows from our calculations it may completely vanish for some transition metals (e.g. Cr—in agreement with the experimental results from [9]).

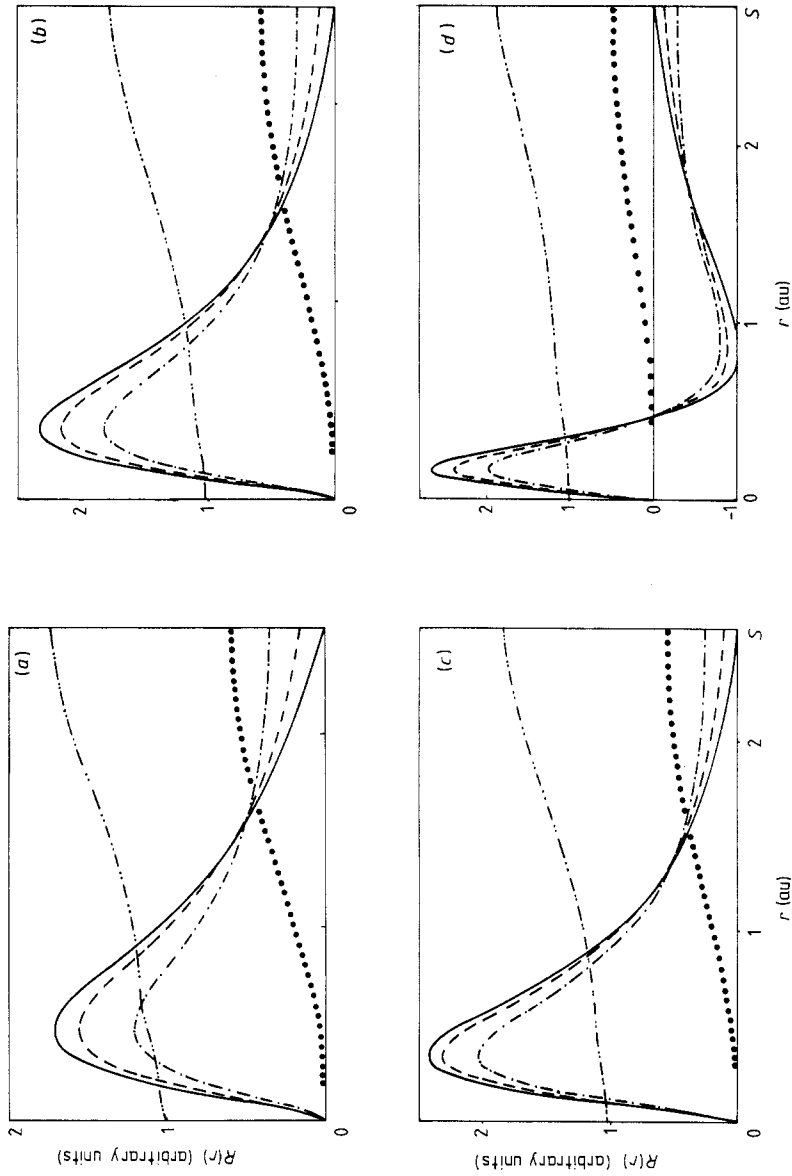
It should be noticed that the model of enhancement based on equations (4) and (5) seems to be general in the sense that it enables us to explain both the relative de-enhancement in Ni and Pd (figure 1)—according to experiment [7, 8] as well as (in some other cases [4, 5, 9]) the increasing enhancement factor with increasing electron energy—also in accordance with experiment.

That is why, in our opinion, such a model of enhancement seems to be most efficient, being physically transparent and, at the same time, quite simple.

The author is grateful to Professor H Stachowiak for various interesting and enlightening discussions.



**Figure 1.** The effective enhancement  $B_2(E_{k,n})$  of d-band transition metals as a function of the energy [12] according to equation (5). (a) Cr, (b) Ni, (c) Cu and (d) Pd with  $X_{k,n}(r) = [E_{k,n} - V(r)]/[E_F - V(r)]$  (---);  $X_{k,n}(r) = E_{k,n}/E_F$  (- · - · -);  $X_{k,n}(r) = 0$  (—).



**Figure 2.** Radial variation of the enhancement  $\{\epsilon[r_s(r), 0]\}^{1/2}$  (—, - - - - -); positron wavefunction (· · · · ·); d-electron wavefunction at the bottom (— · — · —); at the centre (— · — · —); and at the top (—) of the d band within the Wigner-Seitz sphere in (a) Cr, (b) Ni, (c) Cu and (d) Pd.

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- [6] Rubaszek A, Stachowiak H, Boroński E and Szotek Z 1984 *Phys. Rev. B* **30** 2490—actually we have used slightly modified values of the  $\epsilon$ , because those obtained by Rubaszek and co-workers seem to be too strongly increasing near  $p_F$  (Stachowiak H and Boroński E private communication), i.e. starting from  $p = 0.8p_F$  the results have been extrapolated up to  $p = p_F$  by the parabola.
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