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LETTER TO THE EDITOR

Scattering and trapping of positrons at vacancies in solids

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Abstract. Calculations of the positron trapping efficiency at vacancies and their clusters in the transition limit within the optical potential approach are presented. The results are compared with the results of the approach based upon Fermi's golden rule reported in the literature. No significant differences are found either for metals or semiconductors. The calculations show that the elastic scattering of positrons at vacancies and their clusters is more important than trapping. The absorption coefficients for positrons at vacancies and their clusters in aluminium and silicon are evaluated.

The experimental results on the positron lifetime and Doppler broadening of the annihilation line spectroscopy used to be explained by the trapping model that gives the rate equations for the positron annihilation in the delocalized state (as free positrons) and in the localized bound states (as trapped positrons in the vacancy and other open volume defects [1, 2]). The parameters of the trapping model are the positron lifetimes in free and trapped states and the trapping rates which characterize the transition of positrons from the delocalized to the localized states. In the trapping model the trapping rate is assumed to be proportional to the concentration of the defects which capture positrons. The proportional factor is called the specific trapping rate or trapping efficiency related to the cross-section for the trapping process.

The positron lifetimes have been accurately established from experiments and there exist also reliable and practical theoretical methods for finding them, e.g. [3, 4, 5, 6]. The situation is quite the reverse for the trapping efficiency. In that case the experimental values are only poorly estimated. This is related to the uncertainties in the determination of the defect concentration and an actual sample often has defects of several kinds. Only in limited cases (aluminium [7], silicon [8]) has the dependence of the trapping efficiency on temperature been reported. In one case it was also reported that the number of impurity atoms surrounding the vacancy has an effect on the trapping efficiency [9]. Nevertheless, within the last few years the understanding of the positron trapping process and the theoretical estimations of the trapping efficiency have improved.

In the case of vacancies or small vacancy clusters positron trapping is limited by the transition process and the diffusion does not play any important role. The present article is a presentation of theoretical calculations of the trapping efficiency in the transition-limited process, based on the optical potential associated with the vacancy.

The first calculations of the positron trapping efficiency for the vacancy in aluminium were performed by Hodges [10] who considered the electron–hole excitation as a possible mechanism for the energy transfer in the trapping process. In the calculations he adopted Fermi's golden rule. Nevertheless, in his calculations and in the following estimations of the positron trapping efficiency done by other authors the positron wave function was used in

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an approximate way. It led to a weak dependence of the trapping efficiency on temperature, or even its complete absence. That was preferable for positron annihilation experiments, where all detected changes of positron lifetimes and their intensities could be interpreted as a change in the vacancy concentration or in their environment. In the calculations performed by McMullen and Stott [11] the exact analytic positron wave function, both in a trapped and in a free state, was taken into account. This was possible because they modelled the vacancy potential by a square well. Following that method Puska and Manninen [12] and Puska *et al* [13] performed calculations of positron trapping efficiency for vacancies and their clusters in aluminium and in silicon, respectively. The calculations exhibited a strong dependence of the trapping efficiency on temperature or on the energy of the untrapped positron and its resonance behaviour as well. In the calculations based on Fermi's golden rule it is possible to introduce the mechanism of the energy transfer from the positron to the host. In metals it seems that the main mechanism is the electron–hole excitation, but in semiconductors one can also find another mechanism associated with the phonon creation and the excitation of electrons localized at the defect-associated levels. Generally, it is difficult to establish all the possible positron energy loss processes which lead to the trapping of positrons into the vacancy, because there is still no experimental technique to detect the details of those processes. It also seems that the Fermi golden rule (or the Born approximation) is not such a good approximation when the incident energy is small in comparison with the potential energy, e.g. for thermal positrons interacting with defects in solids. In that case it would be interesting to treat the positron trapping process at a vacancy using the optical potential approach. At least at the beginning we can consider the phenomenology approach in which the imaginary part of the optical potential associated with the vacancy is taken as a parameter. This parameter contains all possible energy loss processes. The real part of the potential is the real potential felt by the positron in the vicinity of the vacancy. The description of the positron trapping process as an absorption of the positron wave function at the complex potential associated with the vacancy was first suggested by Shirai and Takamura [14] and then developed by Dryzek [15]. The calculations showed that such an approach can be very useful in the description of the dependence of the trapping efficiency on temperature. The aim of this paper is to show that the optical potential approach produces results similar to those of the approach with Fermi's golden rule, both in metals and in semiconductors. Moreover, it points out the fact that the elastic scattering of the positron at the vacancy is also significant.

We adopted the standard formalism of the quantum theory of scattering for description of the positron trapping process [16]. In the case of metals, where the potential originated from the vacancy is effectively screened by the conducting electrons, one can assume that the vacancy potential is described as a square well:

$$V(r) = \begin{cases} V_0(1 + \mathrm{i}\beta) & \text{for } |r| \leqslant R \\ 0 & \text{for } |r| > R \end{cases} \tag{1}$$

where β is an added extraordinary parameter called the absorption coefficient, V_0 is the real potential felt by the positron in the vicinity of a vacancy and $\mathrm{i} = \sqrt{-1}$. (We also assumed that the absorption coefficient is a constant value; in general the optical potential can be energy dependent and nonlocal.) The depth of the potential V_0 consists of the term with the Coulomb potential (only in the vicinity of the vacancy), the terms with the electron–positron correlation energy and the term coming from the shift of the potential inside the vacancy relative to the potential in the perfect host, the so-called 'zero-shift energy'. In many self-consistent calculations the positron binding energy in the vacancy was estimated. Knowing the value of that energy one can easily get the value for V_0 which causes the

positron bound state. In our calculations we adopted that method to estimate the value V_0. The next step is solving of the Schrödinger equation for the scattering state with the square well potential (1). In a potential symmetric about the z axis, the positron wave function in the scattering state (positive energy) has the form:

$$\psi_k(r) = \frac{1}{kr}\sum_{l=0} u_l(r)Y_{l0}(\Theta) \tag{2}$$

where $k = |k|$ is the positron wave vector, $Y_{l0}(\Theta)$ are the spherical harmonics, $u_l(r)$ is the solution of the radial Schrödinger equation ($l = 0, 1, \ldots$). It can be evaluated in the form:

$$u_l(r) = \begin{cases} c_l K r j_l(Kr), & \text{for } |r| \leqslant R \\ kr[a_l h_l^-(kr) + b_l h_l^+(kr)] & \text{for } |r| > R \end{cases} \tag{3}$$

where $h_l^{\pm} = j_l \pm \mathrm{i}n_l$ are the Hankel functions, and

$$K = \sqrt{\frac{2m^*}{\hbar^2}[E - V_0(1 + \mathrm{i}\beta)]}$$

Also, m^* is the positron effective mass, c_l is a constant and $\hbar$ is the Planck constant divided by 2π. For the description of the absorption and the elastic scattering process we need the scattering matrix which is defined as follows:

$$S_l \equiv \frac{b_l}{a_l} = \exp(2\mathrm{i}\delta_l)$$

where δ_l is the phase shift between outgoing and ingoing positron waves. The scattering matrix can be easily obtained if we take into account the condition that both the positron wave function (3) and its derivative must be continuous at $r = R$. As a result of this we have

$$\exp(2\mathrm{i}\delta_l) = \frac{kj_l(KR)h_{l-1}^-(kR) - Kj_{l-1}(KR)h_l^-(kR)}{Kj_{l-1}(KR)h_l^+(kR) - kj_l(KR)h_{l-1}^+(kR)}. \tag{4}$$

The positron trapping efficiency is associated with the cross-section for the absorption σ_{abs} as follows:

$$\mu(k) = N_{at}\frac{\hbar k}{m^*}\sigma^{abs} = N_{at}\frac{\pi\hbar}{m^*k}\sum_{l=0}^{l_{max}}(2l+1)\left[1 - |\exp(2\mathrm{i}\delta_l)|^2\right] \tag{5}$$

where N_{at} is the atomic density. It was established that

$$l_{max} \approx 2.4kR. \tag{6}$$

In the case of thermalized positrons in a solid l_{max} is close to unity, so only 's', 'p' and/or 'd' partial waves are important in (5). The positron is present in the thermalized state before the trapping process. This shows that in a real solid we have to average the trapping efficiency (5) with the Boltzmann–Maxwell distribution as follows:

$$\mu(T) = \frac{2\hbar^3}{\sqrt{2\pi(k_B T m^*)^3}}\int_0^{\infty} \mathrm{d}k\, k^2\mu(k)\exp[-\hbar^2k^2/2k_B T m^*] \tag{7}$$

where k_B is the Boltzmann constant. In the approach based on the optical potential one can immediately obtain a cross-section for the elastic scattering of the positron at the vacancy. Let us define the scattering efficiency in a similar way as the trapping efficiency, replacing in (5) the cross-section for absorption by the cross-section for the elastic scattering:

$$\nu(k) = N_{at}\frac{\hbar k}{m^*}\sigma^{el} = N_{at}.\frac{\pi\hbar}{m^*k}\sum_{l=0}^{l_{max}}(2l+1)|1 - \exp(2\mathrm{i}\delta_l)|^2. \tag{8}$$

(Note that we have neglected in the calculations inelastic scattering of the positron; this is possible because the energy of the untrapped positron is three orders smaller than the depth of the potential.)

Table 1. Positron binding energy (E_b), and parameters of the square well of the optical potential seen by a positron at the vacancy and its clusters used in the calculations.

Defect	Type of bound state	E_b (eV)	R (Å)	V_0 (eV)	β
Vacancy	s	2.91	1.582	−9.90	0.047
Two-vacancy cluster	s	4.86	1.990	−9.90	0.024
	p	0.17			
Three-vacancy cluster	s	5.55	2.279	−9.85	0.12
	p	1.51			
Four-vacancy cluster	s	6.21	2.526	−9.90	0.04
	p	2.63			

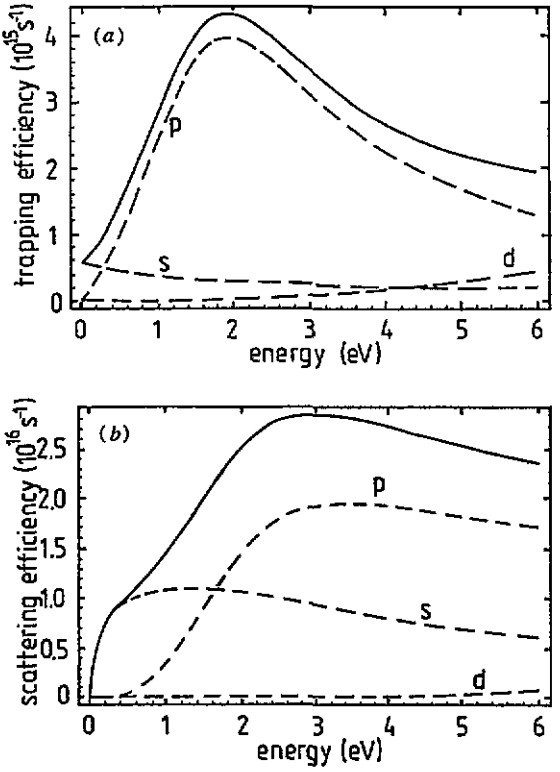

Figure 1. The trapping (a) and scattering (b) efficiency at an aluminium vacancy as a function of the energy of the untrapped positron. The solid lines are the total trapping (5) and scattering (8) efficiency and the dashed lines are the 's', 'p' and 'd' components.

We performed the calculations of the trapping and scattering efficiency for positrons at vacancies and at their clusters in aluminium. In table 1 one can find the potential parameters (V_0, β and R) and the positron binding energies used in the calculations. The

binding energies are close to those used by Puska and Manninen [12] in their calculations. This allows one to compare the results obtained in two methods. We can see the calculated trapping efficiency (5) at a vacancy in aluminium (figure 1(a)) as a function of the untrapped positron energy ($\hbar^2k^2/2m^*$) which is almost the same as the dependence obtained by Puska and Manninen [12] by applying Fermi's golden rule (figure 2 in [12]). In our case we assumed the absorption coefficient β to be equal to 0.047. In both cases resonance of the 'p' partial wave at 1.9 eV is predicted. In figure 1(b) we can see that the scattering efficiency of the positron at the vacancy (8) rapidly decreases when the positron energy is falling to zero. Note that the scattering efficiency is within one order higher than the trapping efficiency. Doing the calculations of the trapping efficiency for vacancy clusters, with the potential parameters presented in table 1, one can obtain almost the same energy dependence as was obtained by Puska and Manninen [12] (figure 6 and figure 8 in [12]). In the case of the temperature dependence the two sets of results are very similar as well. In figure 2 the dependence of trapping (a) and scattering (b) efficiency are presented as a function of temperature. (Figure 2(a)) is similar to figure 9 in [12].) For the vacancy and the vacancy clusters consisting of three and four vacancies, the trapping efficiency slowly increases simultaneously with the increase of temperature. For the divacancy the rapid increase is a result of the resonance trapping of positrons at 0.16 eV. For positrons with thermal energies we are only able to see a tail of the resonance. For decreasing temperature the scattering efficiency decreases both for the vacancies and for their clusters. Still, the scattering efficiency is one order higher than the trapping efficiency. This shows that the elastic scattering of positrons at the vacancies and their clusters is much more important than trapping. At present, there are no direct observations of that phenomenon which could support the validity of our calculations. Nevertheless, the development of experiments with a positron slow beam where the diffusion of positrons towards the surface of a solid is detected could mean that the scattering of positrons at defects could be measured.

The dependence of the trapping efficiency on temperature is extremely sensitive to the potential parameters. In the calculations performed by Puska and Manninen and in our calculations presented in figure 2, the potential parameters did not change along with the temperature. That assumption is difficult to defend because the temperature influences the crystal host causing, e.g., the lattice expansion. The depth of the square well V_0 does not depend on the temperature. But we can assume that the defect radius R is a linear function of temperature: $R(T) = R(1 + \alpha T)$, where α is a linear coefficient. If we take as α the linear expansion coefficient for aluminium and the calculations of the trapping efficiency as a function of temperature (7) are again performed, we get the dependence presented in figure 3(a). Now the resonance dependence of the trapping efficiency on the divacancy is much more visible. In the case of the positron annihilation it was established that e.g. the positron lifetime in bulk increases with temperature linearly but faster than lattice expansion. For aluminium the linear coefficient for the positron bulk lifetime is equal to 1.7×10^{-4} K^{-1} (in [17]). In figure 3(b) we can see the dependence of the trapping efficiency on temperature assuming that value as α. The resonance trapping for the divacancy at 200 K and the fast increase of trapping efficiency for the vacancy and the vacancy clusters with temperature is now obtained. In the literature that rapid increase of the annihilation line shape parameter (S parameter) with temperature for aluminium and aluminium alloys was reported [18] at temperatures close to the melting point. Probably these increases in the vacancy saturation region are the result of the strong dependence of the positron trapping efficiency on temperature, e.g. presented in figure 3(b). The resonance behaviour of the positron's trapping efficiency at the divacancy results from changing of its radius.

As was mentioned, in the literature there have been reported different values for the

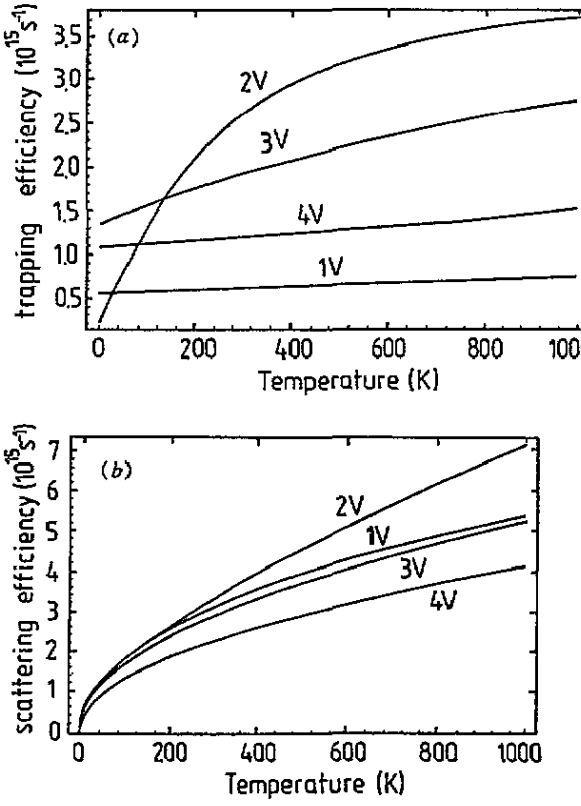

Figure 2. The temperature dependence of the positron trapping (a) and scattering (b) efficiency at the vacancy (1V) and its clusters consisting of two (2V), three (3V) and four (4V) vacancies in aluminium. (No thermal expansion of the lattice was assumed.)

binding energy of a positron in the vacancy, e.g. in the aluminium. We accepted the value −2.3 eV in order to compare our results with the results obtained in [12]. Nevertheless, the two-component self-consistent calculations performed by Boroński and Nieminen [19] gave another value of binding energy in the aluminium vacancy which equals −1.59 eV. Modelling their calculations of the vacancy potential by the square well we obtain the depth V_0 which equals −7.865 eV (which gives the same value of the positron binding energy). From the experiment we know that the positron trapping efficiency for the vacancy in aluminium is equal to 5.7×10^{14} s^{-1} (from [17]). From (7) it is easy to deduce that the complex potential for that case equals $-7.865(1 + 0.026i)$ eV. Figure 4(a) presents the calculated temperature dependence of the trapping efficiency assuming a condition that there is no thermal expansion of the lattice ($\alpha = 0$), $\alpha = 2.6 \times 10^{-5}$ K^{-1} and $\alpha = 1.7 \times 10^{-4}$ K^{-1}. However, total changes of the trapping efficiency presented in figure 4(a) are within 2×10^{14} s^{-1} which is too small for detecting details of the predicted dependence from the actual positron measurement techniques. Thus, from the experimental point of view for the single vacancy in aluminium the trapping efficiency is a constant for the temperature up to the melting point. Certainly that conclusion is valid for the self-consistent calculations done by Boroński and Nieminen [19]. Figure 4(b) presents the dependence of the scattering efficiency in that case.

The examples presented above show that the optical potential approach works well in

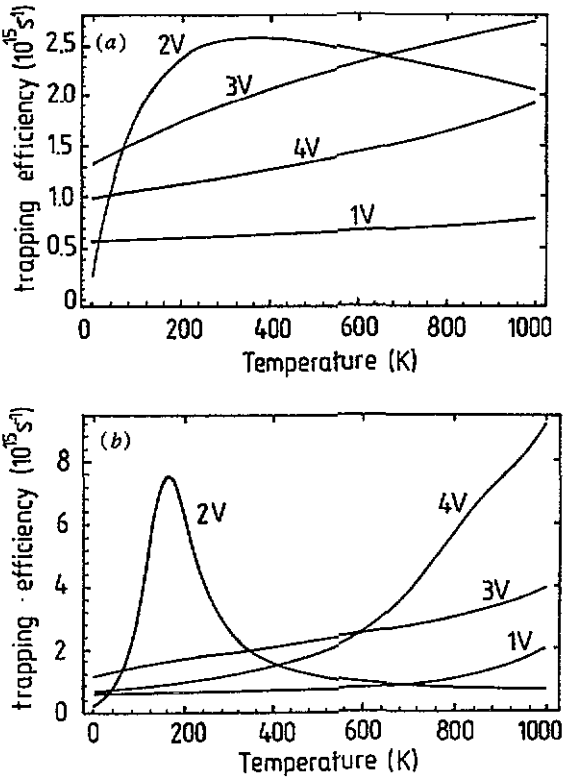

Figure 3. The temperature dependence of the positron trapping efficiency at the vacancy (1V) and its clusters consisting of two (2V), three (3V) and four (4V) vacancies in aluminium. The calculations were performed assuming thermal expansion of the lattice with the linear coefficient equal to $\alpha = 2.6 \times 10^{-5}$ K^{-1} (a) and $\alpha = 1.7 \times 10^{-4}$ K^{-1} (b).

the case of metals. However, one can show that also in the case of semiconductors that approach produces the results close to results obtained by Puska *et al* [13] where Fermi's golden rule was used. In the case of semiconductors the potential which originates from the vacancy and is felt by positrons is not screened by the conduction electrons. Only for neutral vacancies can one assume the potential felt by positrons in their vicinity to be the square well potential (1). The charged vacancies are surrounded by an additional Coulomb potential which is extended over the host. As was shown by Rodriquez *et al* [20] and as was applied by Puska *et al* [13] the potential sensed by the positron at charged vacancies in silicon is as follows:

$$V^{\pm}(r) = \begin{cases} V_2(1 + i\beta_2) & \text{for } |r| \leqslant R_2 \\ V_1(1 + i\beta_1) & \text{for } R_2 < |r| \leqslant R_1 \\ \pm 1/\epsilon_0 r & \text{for } R_1 < |r| \end{cases} \tag{9}$$

where the absorption coefficients β_1 and β_2 were added to our calculations and ϵ_0 is the static dielectric constant. The scattering matrix and the phase shift for that potential are given by

$$S_l \equiv \exp(2i\delta_l) = \frac{A_l + B_l G_l}{C_l + D_l G_l} \tag{10}$$

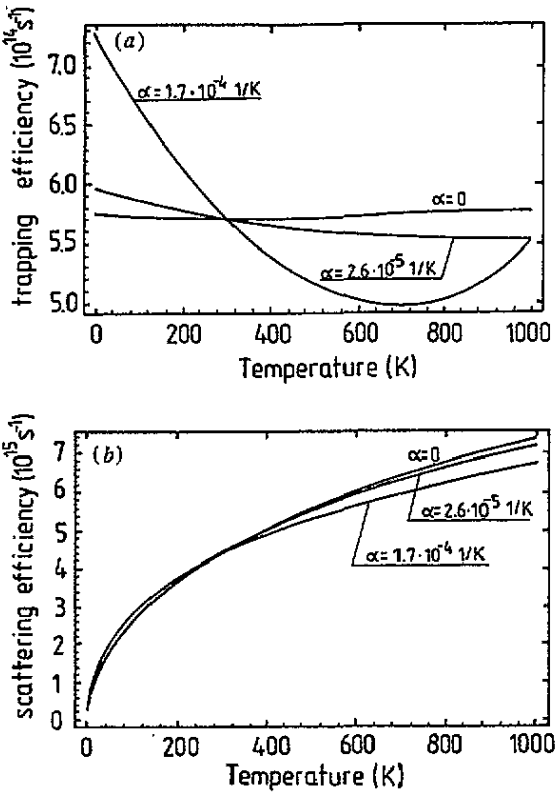

Figure 4. The temperature dependence of the trapping (a) and scattering (b) efficiency for the potential equal to $-7.865(1 + 0.026i)$ eV which modelled the potential obtained in the two-component self-consistent calculations performed by Boroński and Nieminen [19]. The three values of the linear coefficient of the lattice marked in the figure were assumed.

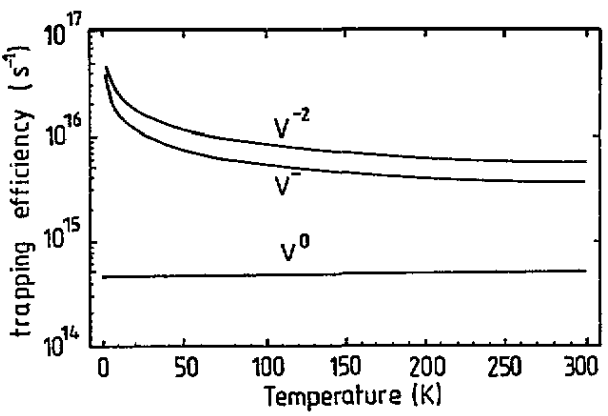

Figure 5. The trapping efficiency for negatively charged and neutral vacancies in silicon; the values of the potential parameters are described in the text.

where

$$G_l = \frac{k_2 j_{l-1}(k_2 R_2) h_l^-(k_1 R_2) - k_1 j_l(k_2 R_2) h_{l-1}^-(k_1 R_2)}{k_1 j_l(k_2 R_2) h_{l-1}^+(k_1 R_2) - k_2 j_{l-1}(k_2 R_2) h_l^+(k_1 R_2)}$$

$$A_l = kh_l^-(k_1R_1)[H_l^-(k, R_1, n/k)]' - k_1[h_l^-(k_1R_1)]'H_l^-(k, R_1, n/k)$$
$$B_l = k[H_l^-(k, R_1, n/k)]'h_l^+(k_1R_1) - k_1H_l^-(k, R_1, n/k)[h_l^+(k_1R_1)]'$$
$$C_l = k_1[h_l^-(k_1R_1)]'H_l^+(k, R_1, n/k) - kh_l^-(k_1R_1)[H_l^+(k, R_1, n/k)]'$$
$$D_l = k_1[h_l^+(k_1R_1)]'H_l^+(k, R_1, n/k) - kh_l^+(k_1R_1)[H_l^+(k, R_1, n/k)]'.$$

$k_{1,2} = \sqrt{(2m^*/\hbar^2)[E - V_{1,2}(1 + i\beta_{1,2})]}$, $H_l^{\pm}(k, r, n/k) = f_l(k, r, n/k) \pm ig_l(k, r, n/k)$ (f_l and g_l are regular and irregular Coulomb functions), $n = \text{sign}(V_1)e^2m^*/(\hbar^2\epsilon_0)$ and e is the electron charge (the prime denotes the derivative with respect to the radial distance r for Coulomb functions and with respect to the argument of the Hankel functions). The following values of the potential parameters were taken in our calculations: $V_2 = -3.6$ eV, $R_2 = 2.540$ Å (for the neutral, singly and doubly negative vacancy), $V_1 = -0.1, -0.2$ eV, $R_1 = 7.1968$ Å (for the singly and doubly negative vacancy, respectively) and $\epsilon_0 = 11.7$, which are the same as in [13]. Taking $\beta_1 = 0.047$ and $\beta_2 = 0.03$ for the singly negative vacancy, $\beta_1 = 0.0235$ and $\beta_2 = 0.03$ for the doubly negative vacancy and $\beta_2 = 0.025$ for the neutral vacancy we get the temperature dependence of the trapping efficiency (7) for the negatively charged and neutral vacancy in silicon presented in figure 5. The dependences obtained are very close to those obtained by Puska *et al* [13] (figure 7 in [13]). One can also achieve a good agreement between the calculation performed by Puska *et al* [13] and the calculations based on the optical model for a positive vacancy. In this case $V_1 = 0.1(1 - 0.12i)$ eV and $V_2 = -3.6(1 + 0.12i)$ eV must be taken in the calculations of trapping efficiency. The calculations show that in semiconductors the optical potential can also be applied successfully to the description of the positron trapping process also at the charged vacancies.

Summing up one can point out that the positron trapping efficiency at the vacancies and their clusters is a function of the untrapped positron's energy or of temperature. The methods of calculation of the trapping efficiency based on the Fermi golden rule and on the optical potential generally produce very close results both in metals and in semiconductors. However, the calculations based on the optical potential indicated that the process of elastic scattering of positrons on the vacancy and its clusters is much more efficient than the process of trapping. The optical potential allows us also to introduce the spatial localization of the positron loss energy process which will be examined more carefully in our further calculations. At present the experimental documentation on the trapping rate and the trapping efficiency is still poor, and the evaluation of the results of calculations is rather qualitative.

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