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The optical potential responsible for the absorption of a positron at a vacancy

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Abstract. We describe an approach to the positron trapping at vacancies based on the optical theorem. We calculate the imaginary part of the optical potential in an *ab initio* model at the time when electron–hole excitation takes place. The obtained analytical formulae for the optical potential are tested as a function of the positron incident energy, the positron binding energy, and the electron density parameter r_s. The calculation of the optical potential range agrees well with the vacancy radius. The approach is used for the description of observed temperature dependences of trapping rate at vacancies in pure metals as reported in the literature. The results are satisfactory and the obtained values of the optical potential are quite reasonable.

1. Introduction

The inelastic scattering and the trapping of positrons at vacancies in a solid are characterized by the trapping rate efficiency parameter. This parameter and positron mean lifetime are the only parameters in the rate equations, called the trapping model [1], which is used in the description of the fate of positrons in a solid. The mean positron lifetime at a vacancy is well understood from the theoretical point of view and it is easy to measure. The trapping rate efficiency parameter, which describes the transition rate from the free to the trapped state, is rather poorly estimated from experiments. We can add that in the problem of the trapping and scattering of positrons by vacancies in solids, an exact theoretical treatment involving the interactions between the incident positron and all the electrons is complicated. Nevertheless, the proper description of these processes could be helpful in a better understanding of the vacancy structure and its electronic environment detected by positrons. There are also experimental proofs that the trapping efficiency parameter depends on the vacancy surroundings and it exhibits a temperature dependence.

In the calculation of the trapping efficiency the theory of scattering and trapping of thermal neutrons at nuclei [2] was included. Authors who apply this theory take the simple relation for the cross section of neutron absorption at nuclei and try to describe the absorption of positrons at vacancies, e.g. in [3]. However, the relations which are valid for neutrons are not valid for positrons. This is due to the fact that in this theory it is assumed that the neutron incoming wave is attenuated in the region of the nucleus but can still be described as a plain wave. We know from other authors, e.g. [11], that the trapped positron is well localized, and its wave function looks more like a Gaussian function than a plane wave. The authors [3] consider only the absorption of the ‘s’ wave but it is easy to show that at higher temperature the ‘p’ wave also gives an important contribution.

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A better approach to the problem of calculating the trapping efficiency came from Hodges, who applied the Fermi golden rule [4], but this has severe limitations since it has to be combined with an *ad hoc* assumption on how the transition occurs [5].

Here we present the approach to trap positrons at vacancies by the scattering theory with the optical potential first suggested by Shirai and Takamura [6]. This approach seems to be a much better description of the problem of positron trapping, since it takes care of the elastic and inelastic scattering of the positron wave function. The value of the optical potential can either be calculated from theory or deduced from experiment. The basic idea is that we consider the scatter of a positron at a vacancy immersed in the electron sea. The positron will be scattered, either elastically or inelastically. Instead of treating the interaction of a positron with each individual electron one seeks an 'effective' potential between positron and vacancy, such that the scattering by the potential describes the observed effect. As shown by Feshbach [7], in many cases the optical potential successfully describes the properties of the cross section of the scattering of neutrons at nuclei. The optical potential is frequently used the in description of nuclear reaction data but also for low-energy positron or electron diffraction data.

We intend to present an outline of the theory of positron scattering at a vacancy and the method of calculation of the imaginary part of the optical potential *ab initio* for the electron–hole excitation. The application of the approach to the description of data reported in the literature will also be presented.

2. The optical model

In our consideration we take into account that the trapped positron loses its energy in a process of creation of holes and electrons in the excited state above the Fermi level. We consider the interaction of an incident positron with an electron located close to the vacancy. The Schrödinger equation of such a system (figure 1) is given by ($\hbar = 1$)

$$\left(-\frac{1}{2m}\nabla_1^2 + V^+(\boldsymbol{r}_1) - \frac{1}{2m_e}\nabla_2^2 + V^-(\boldsymbol{r}_2) + V(\boldsymbol{r}_1, \boldsymbol{r}_2)\right)\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2) = (E + E_0)\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2) \tag{1}$$

where $V^+(\boldsymbol{r}_1)$ represents the potential of the interaction between positron and vacancy, $V^-(\boldsymbol{r}_2)$ that between electron and vacancy and $V(\boldsymbol{r}_1, \boldsymbol{r}_2) = e^2/|\boldsymbol{r}_1 - \boldsymbol{r}_2|$ between an electron and an incident positron; e is the electron charge, and m and m_e are the positron and electron mass, respectively. $V^-(\boldsymbol{r}_2)$ will be neglected in the following consideration. The momentum of the incident positron is equal to $\boldsymbol{p}$ and the initial momentum of an electron is $\boldsymbol{k}_0$, $E = p^2/2m$ and $E_0 = k_0^2/2m_e$. The wave function of the system $\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2)$ can be

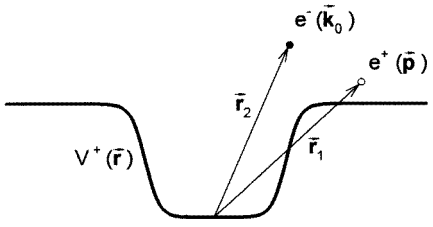


Figure 1. The scattering of a positron in the vicinity of a vacancy, assuming that the inelastic process is realized by electron–hole excitation.

expressed by a 'coupled state' which is the expansion in terms of a set of unperturbed states of the electron $\phi_j(\boldsymbol{r})$:

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2) = \sum_{i=0} \phi_i(\boldsymbol{r}_2)\psi_i(\boldsymbol{r}_1) \tag{2}$$

where $\psi_0(\boldsymbol{r}_1)$ describes the elastic scattering positron wave. As mentioned above we can solve instead of (1) the following equation:

$$\left(-\frac{1}{2m}\nabla^2 + V^+(\boldsymbol{r}) - \frac{p^2}{2m} + \tilde{U}(\boldsymbol{r})\right)\psi_0(\boldsymbol{r}) = 0 \tag{3}$$

where $\tilde{U}(\boldsymbol{r})$ is the so-called 'optical potential'. We adopted the approach suggested by Shaw [8], with the optical potential expressed by

$$\tilde{U} = \langle p|R|p\rangle \tag{4}$$

where R is the operator defined as

$$R = V + \lim_{\eta\to 0} V\frac{1}{E - H_0 + \mathrm{i}\eta}R \tag{5}$$

where $H_0 = -1/(2m)\nabla^2$. In general the optical potential is energy dependent, nonlocal and complex. Nevertheless, in many applications it is enough to assume that the potential is constant in the vacancy range but complex: $\tilde{U} = U - \mathrm{i}W$. In our previous work, [9] we presented the exact solution of (3) for the case of square-well potential V^+ where $W =$ constant but different from zero only within the vacancy region. Note that the trapping efficiency depends directly on the value of the imaginary part of the optical potential as follows [10]:

$$\mu = N_{at}\frac{p}{m}\sigma^{abs} = N_{at}\frac{4\pi\hbar}{Em}\sum_{l=0}(2l+1)\int_0^\infty \mathrm{d}r\, W(r)|u_l(r)|^2 \tag{6}$$

where N_{at} is the atomic density and $u_l(r)$ is the radial solution of (3) which satisfies the integral equation

$$u_l(r) = 2\mathrm{i}kr j_l(kr) + \frac{2m}{k}\int_0^\infty \mathrm{d}r'\, j_l(kr_<)h_l^{(1)}(kr_>)k^2 rr'[V^+(r') + U(r') - \mathrm{i}W(r')]u_l(r') \tag{7}$$

where $r_<$ is the lesser of (r, r'), $r_>$ is the greater of (r, r'), $h_l^{(1)} = j_l + \mathrm{i}\eta_l$ is the Hankel function, and j_l and η_l are the spherical Bessel functions.

In the first-order approximation one can find the relation for the R operator and, hence, the imaginary part of the optical potential [8]:

$$W = \pi\sum_{\boldsymbol{k}}\sum_{\boldsymbol{q}} |M(\boldsymbol{p}, \boldsymbol{q})|^2\Theta(k_F - k)[1 - \Theta(k_F - |\boldsymbol{k} + \boldsymbol{q}|)]\delta(E + E_0 - E_1 + E_b) \tag{8}$$

where $E_b(> 0)$ is the positron binding energy at the vacancy, $E_1 = |\boldsymbol{k} + \boldsymbol{q}|^2/2m_e$ is the energy of the electron in the excited state, k_F is the Fermi momentum, and the matrix element between the initial and final state is

$$M(\boldsymbol{p}, \boldsymbol{q}) = \langle \boldsymbol{k}_0 + \boldsymbol{q}, b|V|\boldsymbol{k}_0, \boldsymbol{p}\rangle \tag{9}$$

where $|b\rangle$ is the positron wave function in the bound state. Further we assume that $\langle \boldsymbol{r}|\boldsymbol{k}_0\rangle = \Omega^{-1/2}\exp(\mathrm{i}\boldsymbol{k}_0\cdot\boldsymbol{r})$, $4\langle \boldsymbol{r}|\boldsymbol{k}_0 + \boldsymbol{q}\rangle = \Omega^{-1/2}\exp[\mathrm{i}(\boldsymbol{k}_0 + \boldsymbol{q})\cdot\boldsymbol{r}]$, also that the incident positron wave can be described as a plane wave, $\langle \boldsymbol{r}|\boldsymbol{p}\rangle = \Omega^{1/2}\exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{r})$, and Ω is the normalization volume. Thus one can rewrite the matrix element of (9) in the form

$$M(\boldsymbol{p}, \boldsymbol{q}) = \Omega^{-3/2}V(q)\int \mathrm{d}\boldsymbol{r}\, \psi_b(\boldsymbol{r})\exp[-\mathrm{i}(\boldsymbol{p} - \boldsymbol{q})\cdot\boldsymbol{r}] \tag{10}$$

where $V(q) = 4\pi e^2/[q^2\varepsilon(q)]$, and $\varepsilon(q)$ is the dielectric function of the host. We assume that the positron in the trapped state is described by a Gaussian function [11]:

$$\psi_b(\boldsymbol{r}) = (2a/\pi)^{3/4}\exp(-ar^2) \tag{11}$$

where a is a parameter. Finally we can rewrite the imaginary potential W as follows:

$$W = \sqrt{\frac{2}{\pi a^3}}\frac{k_F\alpha^2 mc^2}{\Omega}\int_{-1}^{1}\mathrm{d}t\int_0^{\infty}\mathrm{d}q\,\frac{\exp[-(p^2+q^2-2pqt)/(2a)]}{q^2\varepsilon(q)^2} \times K\left(\frac{q}{k_F},\frac{0.5p^2+mE_b}{k_F^2}\right) \tag{12}$$

where $K(x, y) = \pi[1-(y/x-0.5x)^2]/x$ if $x+0.5x^2 \geqslant y \geqslant |x-0.5x^2|$, $K(x, y) = 2\pi y/x$ if $0 \leqslant y \leqslant x-0.5x^2$ and $x < 2$, and $K(x, y) = 0$ otherwise [5], α is the fine-structure constant, and c is the speed of light.

3. Discussion

The imaginary potential depends on the positron binding energy E_b, the Fermi momentum, k_F ($k_F = 0.3169/(r_s/a_0)$ [nm^{-1}], where a_0 is the Bohr radius) and also on the value of momentum, or energy, of the incident positron, $|\boldsymbol{p}|$. The potential depends on the dielectric function, $\varepsilon(q)$. In order to avoid the logarithmic singularity we can take the Thomas–Fermi approximation of the dielectric function, $\varepsilon(q) = 1 + k_{TF}^2/q^2$, where k_{TF} is the Thomas–Fermi wavenumber ($k_{TF} = 0.295/\sqrt{r_s/a_0}$ [nm^{-1}]). Figure 2 presents the calculated value of the imaginary potential, W, as a function of incident positron energy, according to (12). It presents the case of a vacancy in an Al host: this means that it was assumed that $r_s = 2.07$ a_0, $E_b = 1.56$ eV and $a = 0.0819$ nm^{-2} [11]. Since the value of the potential decreases with increasing positron energy we should not expect a trapping of nonthermal positrons.

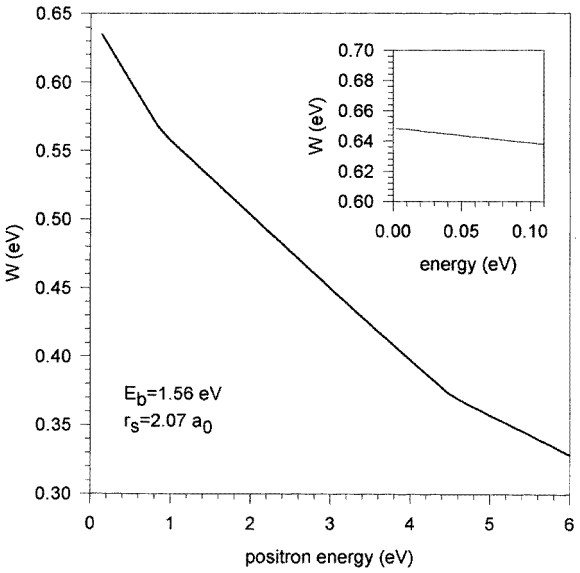


Figure 2. The imaginary part of the optical potential as a function of incident positron energy for $E_b = 1.56$ eV and $r_s = 2.07$ a_0.

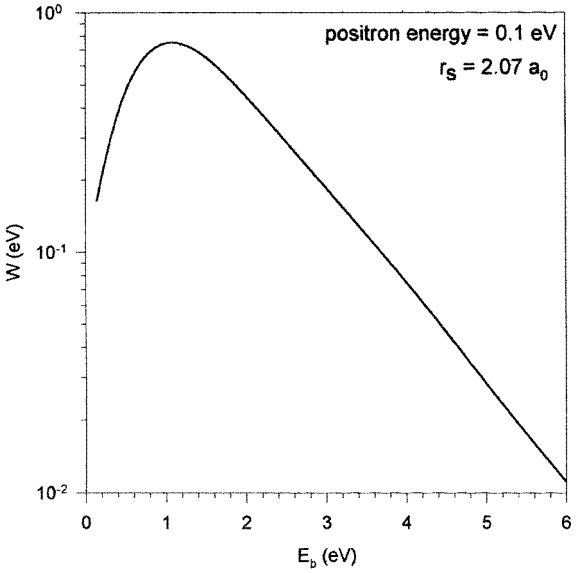


Figure 3. The imaginary part of the optical potential as a function of positron binding energy for $E = 0.1$ eV and $r_s = 2.07\ a_0$.

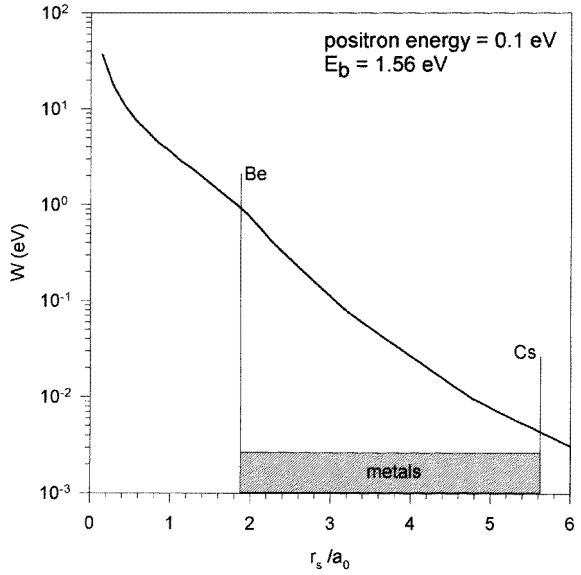


Figure 4. The imaginary part of the optical potential as a function of the r_s parameter for $E_b = 1.56$ eV and $E = 0.1$ eV.

An important result is that in the narrow range of thermal positron energy (i.e. less than 0.1 eV), W is almost constant and equal to 0.64 eV. The imaginary potential increases when the value of the positron binding energy increases but after reaching a maximum it rapidly decreases (figure 3), so for deep positron traps we should not expect larger values of the trapping efficiency than for traps for which the positron binding energy is about 1 eV.

Nevertheless, if for some reason the positron binding energy is changing, this should be reflected in the value of trapping efficiency. It is interesting that the imaginary potential almost exponentially decreases with increase of the r_s parameter (figure 4). For metals the r_s parameter alters from 1.9 to 5.6 a_0 and the calculation shows that this induces a change of W by two orders of magnitude. Thus, for metals with a high r_s parameter (such as Cs, Na, Rb), we do not observe positron trapping at their vacancies, explained by the small value of the corresponding W. However, we should keep in mind that around the vacancy, where a positron is localized, the local r_s parameter may be smaller than in the host.

It is interesting to ask about the range of the optical potential one can calculate. The mean square radius of the optical potential from the relation in [12] is

$$\langle R^2 \rangle = -3\left(\frac{1}{U_0(q)}\frac{\partial^2 U_0(q)}{\partial q^2}\right)_{q=0} \tag{13}$$

where U_0 is the zeroth-order approximation of the optical potential, which from (4) is equal to $V(q)$ and hence

$$\langle R^2 \rangle = 6/k_{TF}^2. \tag{14}$$

For Al, $\sqrt{\langle R^2 \rangle} = 0.12$ nm, that is a little smaller than the vacancy radius of 0.158 nm. Nevertheless, if one takes into account the relaxation effects, the range of the optical potential agrees with that of the vacancy potential felt by a positron, V^+. It is worth noting that the optical potential has also a real part which can modify the V^+ potential used in calculations of the positron lifetime at a vacancy.

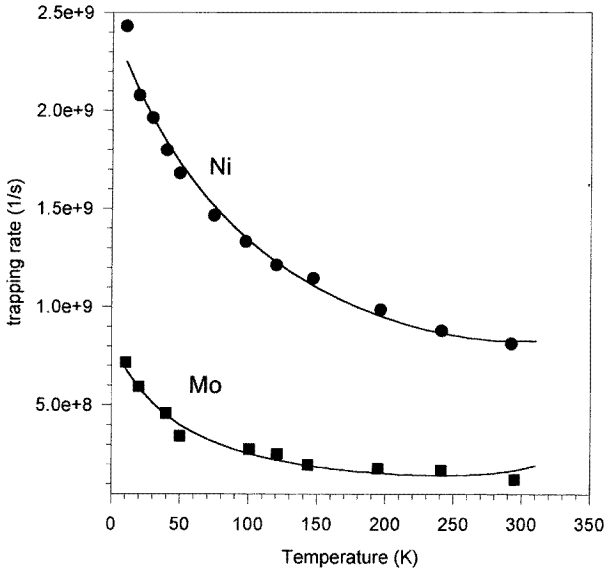
4. Comparison with experimental data

Recently Trump and Petersen [13] performed low-temperature measurements of positron lifetime spectra for single-crystal samples of pure metals: Al, Ni, Zn and Mo. They assumed that the vacancy provides an essential contribution to the spectra and hence they deduced the temperature dependence of the trapping rate of positrons at the vacancy. They pointed out that the trapping rate is proportional to $T^{-1/2}$ over a wide temperature range for the metals studied. That result was explained using the relation on the cross section of absorption of thermal neutrons. As we mentioned above that is a rather crude approach to the positron trapping efficiency. Thus we apply the optical model presented above to describe the data obtained by those authors. For simplification it is assumed that the positron–vacancy interaction in (3) is represented by a square-well potential:

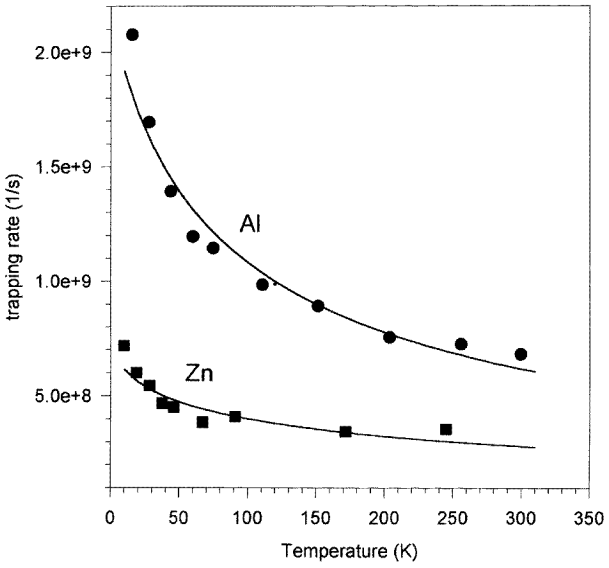
$$V^+(\boldsymbol{r}) + \tilde{U}(\boldsymbol{r}) = \begin{cases} V_0 - \mathrm{i}W & |\boldsymbol{r}| \leqslant R \\ 0 & |\boldsymbol{r}| > R \end{cases} \tag{15}$$

where R represents the vacancy radius. For such a case the solution of (3) may be expressed in analytical form, e.g., (7) in [9]. In this procedure the following parameters were fitted: V_0, W, and the vacancy concentration C. We assumed that the vacancy radius is a linear function of temperature, $R = R_0(1 + \alpha T)$, and R_0 and α were also fitted. This assumption was necessary due to the experimental evidence of the thermal expansion of the vacancy volume [14].

The solid lines in figure 5 present the best fit to the experimental points of the temperature dependence of the trapping rate: $K(T) = C^*\mu(T)$, where $\mu(T)$ is the trapping efficiency with the Boltzmann–Maxwell distribution defined by (7) in [9]. Table 1 contains the values from the fit. First of all, one can find excellent agreement between the presented theory and the experiment for Ni and Mo but rather poor agreement for Zn. It is interesting



(a)



(b)

Figure 5. The temperature dependence of the positron trapping rate at vacancies in Ni, Mo (a), and Al, Zn (b); the experimental points were taken from [13] and the solid lines are the best fit to the calculated trapping rates from the optical potential approach.

that in all cases the vacancy radius obtained from the fit is lower than that obtained from the corresponding atomic volume. This is consistent with the inward relaxation of atoms around the vacancies in metals as detected by other measurements. The depth of the vacancy potential, V_0, evaluated in the fit is in good agreement with other theoretical calculations, e.g. with the value of 7.88 eV found by Boroński and Nieminen [15] for a vacancy in

Al. The imaginary part of the optical potential W obtained from the fit for Al is almost equal to the value obtained from the calculation presented above, according to (12). Note the large value of the thermal coefficient α obtained from the fit. For Al, this value is about a factor of 20 larger than the experimental linear expansion coefficient. This result is not too surprising since the measurements of the self-diffusion activation volumes in Cd indicated a value of the thermal expansion coefficient of the vacancy volume about a factor of 15 larger than the macroscopic volume thermal expansion coefficient [14]. We expect that the atomic vibration may also contribute essentially to the temperature change of the vacancy volume and hence the positron–vacancy potential. This effect may explain the poor agreement between theory and experiment obtained for Zn, of which the Debye temperature is more than 100 K lower than for the other metals studied.

Table 1. The parameters of the optical potential obtained from the fitting procedure.

	R_0 [nm]	V_0 [eV]	W [eV]	C [$\times 10^{-7}$]	α [$10^{-4} \times 1/K$]
Ni	0.136 ± 0.002	-8.12 ± 0.1	0.74 ± 0.01	6.5 ± 0.4	10 ± 2
Mo	0.134 ± 0.001	-7.2 ± 0.04	0.53 ± 0.01	1.34 ± 0.04	16.6 ± 0.4
Zn	0.132 ± 0.01	-8.67 ± 0.5	2.1 ± 0.1	0.81 ± 0.1	5.4 ± 0.04
Al	0.144 ± 0.001	-7.12 ± 0.1	0.62 ± 0.01	4.6 ± 0.3	8.4 ± 0.2

Finally, one can conclude that the optical model is an appropriate approach to calculate trapping efficiencies. The present calculations have shown that the imaginary part of the optical potential related to the electron–hole excitation is responsible for the positron absorption at a vacancy, leading to trapping efficiencies which agree with experiment. The dependence of this potential on the incident positron energy, the positron binding energy and the r_s parameter qualitatively explains the experimentally observed phenomena. The optical model was also able to describe the temperature dependence of the trapping rate reported in the literature. The values of the optical potential obtained from the fit were quite reasonable and agree with other theoretical calculations.

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