Spin polarized positron lifetimes in ferromagnetic metals: First-principles study

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We carry out spin-polarized positron lifetime calculations for ferromagnetic metals using the electron-positron density functional theory (DFT). We investigate Fe, Co, and Ni and find that the differences between the positron lifetimes for their minority and majority spins ($\tau^{\downarrow} - \tau^{\uparrow}$) are 11.85, 3.75, and -4.37 ps, respectively. The negative lifetime difference of Ni is presumed to originate from an unlocalized distribution of minority electrons.

1. Introduction

Recently, spin-polarized positron experiments have attracted scientific attention because of their application to the study of electron spin phenomena.[1,2)] Low-energy spin-polarized positron beams enable us to study magnetism at surfaces and interfaces and in thin films. Positrons are trapped by vacancy defects, and so spin-polarized positron annihilation spectroscopy (SP-PAS) is expected to be a useful tool for studying vacancy-induced magnetism.[3)] Ferromagnets are fundamental spin polarization materials. Therefore, detecting spin polarization in ferromagnets by SP-PAS is scientifically important.

A 3γ spin-polarized positron experiment on ferromagnetic materials was first carried out by Berko and Mills in 1971.[4)] Later, the two-dimensional two-photon angular correlation of spin-polarized positron annihilation radiation in Ni[5–7)] and Co,[8)] and the Doppler broadening of annihilation radiation[1,2)] were experimentally measured. These experiments enabled us to obtain information on spin polarization in ferromagnetic materials. Thus, it is expected that positron annihilation experiment will eventually be applied to the study of spintronics.

There are still only a few theoretical studies of spin-polarized positrons. However, to establish the reliability of SP-PAS, theoretical calculations of spin-polarized positrons are necessary.

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In this study, we carry out calculations of the lifetimes of spin-polarized positrons for the ferromagnetic materials Fe, Co, and Ni. We previously succeeded in carrying out positron-electron density functional theory calculations of positron lifetimes for nonmagnetic systems and well reproduced experimental results.[9,10] In this study, we develop a method of calculating lifetimes in electron spin-polarized systems. Our calculations show that the differences between the positron lifetimes for their minority spin and majority spin ($\tau^{\downarrow} - \tau^{\uparrow}$) are positive for Fe and Co, and negative for Ni. We find that the negative lifetime difference is due to the fact that the distribution of minority spin electrons is rather broad compared with that of majority spin electrons.

2. Calculation methods

In this study, we carry out calculations of positron lifetimes for ferromagnetic metals based on the electron-positron density functional theory (DFT).[11–13] We first perform standard electron band structure calculations based on the spin-polarized generalized gradient approximation. Then, we calculate the positron wave function using the following equation based on the local density approximation for the electron-positron correlation:[12]

$$\{-\frac{1}{2}\nabla^2 - \int dr' \frac{n_e(r') - n_0(r')}{|r - r'|} + \mu_{e-p}(n_e)\}\psi_i^+(r) = \varepsilon_i^+ \psi_i^+(r), \tag{1}$$

where n_e, n_p, and n_0 are the electron, positron, and nuclear charge densities, respectively, and $\mu_{e-p}(n_e)$ is the potential due to the electron-positron correlation. After we obtain the electron ($n_e^{\uparrow(\downarrow)}$) and positron ($n_p$) densities, the positron lifetime $\tau^{\uparrow}(\tau^{\downarrow})$ for the majority (minority) spin can be calculated as

$$\frac{1}{\tau^{\uparrow(\downarrow)}} = \pi r_e^2 c \int dr n_e^{\uparrow(\downarrow)}(r) n_p(r) \Gamma(n_e), \tag{2}$$

where r_e is the classical electron radius and c is the light velocity. $\Gamma(n_e)$ is the enhancement factor.

In the above computational scheme, we assume that the correlation potential $\mu_{e-p}(n_e)$ does not depend on the electron spin-polarization. The calculation of the correlation energy for a single positron in the homogeneous nonpolarized electron gas was performed by the basis of the random phase approximation by Arponen and Pajanne[14] and the numerical results were fitted to an analytic function by Puska, Seitsonen, and Nieminen (PSN)[12] . The enhancement factor $\Gamma(n_e)$ is also deduced from the numerical results of a single positron in a homogeneous non-polarized electron gas. The calculation was performed by Lantto,[15] who used a hypernetted chain method, and the numerical results are fitted to an analytical function by

PSN:[12]

$$\Gamma(n_e) = 1 + 1.23r_s + 0.9889r_s^{3/2} - 1.4820r_s^2 + 0.3956r_s^{5/2} + r_s^3/6, \quad (3)$$

where $(4\pi/3)r_s^3 = 1/n_e$.

The above scheme based on the electron-positron DFT has been applied to various materials and was found to give reliable results for non-spin-polarized systems.[13,16–18] However, applications to the study of spin-polarized materials are very rare. The purpose of this paper is to clarify the validity of the above-mentioned scheme for ferromagnetic materials.

We use the first-principles code PHASE[19] based on the above-mentioned scheme. In the numerical calculations, we adopt ultrasoft pseudopotentials and plane waves. The method of ultrasoft pseudopotentials and plane waves to the calculations of positron lifetimes was presented in our previous paper.[13]

The ferromagnetic metals Fe, Co, and Ni form bcc, hcp, and fcc structures at room temperature, respectively. We use the experimental lattice constants for Fe (2.8675 Å),[20] Co (2.5074 and 4.0699 Å for the a and c lattice constants, respectively), and Ni (3.524 Å).[21] The k-points used in calculations are $60 \times 60 \times 60$, $30 \times 30 \times 18$, and $50 \times 50 \times 50$, for Fe, Co, and Ni, respectively. The cutoff energies of wavefunction are 340 eV for electrons and 680 eV for positrons, and the cutoff energy of charge density is 3040 eV.

3. Results and discussion

First, we investigate the lifetimes of the non-spin-polarized states of Fe, Co, and Ni (Table I). Our calculations well reproduce the experimental lifetimes, although the calculated results are slightly smaller than the experimental results.

Next, we study the spin-polarized states of Fe, Co, and Ni (Table II). The lifetime differences ($\tau^{\downarrow} - \tau^{\uparrow}$) are estimated to be 11.85, 3.75, and -4.37 ps for Fe, Co, and Ni, respectively.

The negative value for Ni is unexpected since the annihilation rate is expected to be large when the electron number is large. However, the negative lifetime difference in Ni is consistent with the previous three-photon polarization measurement,[4] as is discussed here. First, the experimentally observed $P^{3\gamma}$ is given by

$$P^{3\gamma} = \frac{N_+^{3\gamma} - N_-^{3\gamma}}{N_+^{3\gamma} + N_-^{3\gamma}}, \quad (4)$$

where $N^{3\gamma} = \int dp N^{3\gamma}(p)$, $N^{3\gamma}(p)$ is the electron momentum distribution from a three-photon positron annihilation experiment. By using the experimental values of $N_{\pm}^{3\gamma}$, we evaluate $P^{3\gamma}$,

which is denoted by $P^{3\gamma}_{expt}$ in Table III. $P^{3\gamma}$ is also given by

$$P^{3\gamma} = -\frac{(\tau^{\downarrow} - \tau^{\uparrow})(\tau^{\downarrow} + \tau^{\uparrow})}{\tau^{\downarrow 2} + \tau^{\downarrow}\tau^{\uparrow} + \tau^{\uparrow 2}} P^{p} \tag{5}$$

where $\tau^{\uparrow}$ and $\tau^{\downarrow}$ are the positron lifetimes for the majority and minority spins, respectively, and P^{p} is the positron polarization. We evaluate $P^{3\gamma}$ in Eq. (5) using the calculated positron lifetimes ($\tau^{\uparrow}$ and $\tau^{\downarrow}$) and the experimental P^{p} (0.35).[22] This evaluated value is denoted by $P^{3\gamma}_{theory}$ in Table III. Our calculated values ($P^{3\gamma}_{theory}$) are comparable with the experimental values ($P^{3\gamma}_{expt}$), although the theoretical absolute value of Fe is 5.1 times larger than the experimental value. The difference may be due to some inaccuracy in the experiment. It should be emphasized that the signs of the theoretical values of $P^{3\gamma}$ are the same as those of the experimental ones. The positive value of Ni indicates the negative positron lifetime difference ($\tau^{\downarrow}$-$\tau^{\uparrow}$).

Here, we analyze electronic structures to determine the reason for the negative lifetime difference in Ni. We calculate the projected density of states (PDOSs) for the atomic orbitals of 3*d* and 4*s* with a radius of 1.5 *bhor* (Fig. 1). In the cases of Fe, Co, and Ni, the 4*s* components are small compared with the 3*d* components. We also find that the distributions of the PDOSs of *d* orbitals in the three materials are similar to those of the total DOSs [see Figs. 2(a) -2(c)], indicating that the *d* orbital components are much larger than the *s* orbital components. Therefore, 3*d* electrons are considered to dominate the positron lifetime.

We find that the magnetic moment decreases as the atomic number increases: the calculated magnetic moments per atom are 2.311, 1.637, and 0.945 μB, for Fe, Co, and Ni, respectively (see Table II). As the magnetic moment decreases, the difference between the numbers of the majority spin and minority spin electrons becomes smaller, and thus the lifetime difference ($\tau^{\downarrow}$-$\tau^{\uparrow}$) is also expected to become smaller. This expectation is consistent with our calculational results that the positron lifetime difference becomes smaller as the atomic number increases (Table II).

However, the negative lifetime difference in the Ni case is still unresolved. Here, we analyze the electron and positron distributions of Fe and Ni and discuss the negative lifetime difference of Ni. In the case of Ni, the density of the minority spin is delocalized and thus the densities in the interstitial regions are larger than those of the majority spin (Figs. 3) It is found that the density of the positron is localized in the interstitial regions [Fig. 3(d)]. Since the density of the minority spin is large in the interstitial region, the overlaps S_{e-p} are larger

than those for the majority spin [Figs. 3(f) and 3(g)], where S_{e-p} is defined as

$$S_{e-p} = n_e^{\uparrow(\downarrow)}(r) n_p(r) \Gamma(n_e) \tag{6}$$

This large electron-positron overlap for the minority spin is the reason for the negative lifetime difference.

In the case of Fe, we also find that the minority spin electrons have a broad distribution and thus the densities in the interstitial regions are larger than those for the majority spin [Figs. 4(a) and 4(b)]. The positron density is maximum in the interstitial region, as in the case of Ni [Fig. 4(d)]. However, the electron-positron overlap distribution is quite different from that of Ni: since the electron spin polarization is large in the case of Fe, the electron-positron overlap for the majority spin is generally larger than that for the minority spin (Fig. 4). This result leads to the fact that the lifetime difference is positive in the case of Fe.

We also find that the density of the minority spin in the case of Co is rather broad compared with that of the majority spin, and that the positron is localized in the interstitial region. Since the magnetic moment of Co is large, the lifetime difference is positive, as in the case of Fe.

Thus far, we have found that the distribution of the minority spin is broad for Fe, Co, and Ni. We also found that the positron distributions have maxima in the interstitial regions. As a result, the lifetime difference of Ni is negative. However, in the cases of Fe and Co, the lifetimes differences are positive since the magnetic moments are large.

4. Conclusions

In this study, we carried out calculations of the lifetimes of spin-polarized positrons in Fe, Co, and Ni using the electron-positron DFT. First, we calculated the non-spin-polarized positron lifetimes and found that they are close to the experimental values. In the spin-polarized positron lifetime calculations, we found that the lifetime differences ($\tau^{\downarrow} - \tau^{\uparrow}$) are 11.85, 3.75, and -4.36 ps for Fe, Co, and Ni, respectively. The positive and negative values for Fe and Ni are consistent with the results of the 3γ experiment. It is suggested that the negative lifetime difference for Ni originates from the unlocalized behavior of minority electrons. We expect that when the magnetic moment of the material is small, the negative lifetime difference tends to be observed since an unlocalized behavior of minority electrons is expected in other systems.

The agreement between the results obtained using the theory and the experiment suggests that the presently used calculational scheme is reliable for determining the lifetimes

of spin-polarized positrons in electron spin-polarized materials. Therefore, the present calculational scheme is expected to be an effective tool for analyzing the experimental lifetimes. The observation of the momentum distribution of the spin-polarized positrons[1,2] as well as the lifetime measurement emerges as a powerful tool for studying spintronics. It is expected that the present calculational scheme is also useful for analyzing the momentum distribution, although further theoretical study is necessary to confirm the reliability of the theory for the momentum distribution.

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Table I. Non-spin-polarized positron lifetimes (ps) of bulk Fe, Ni, and Co.

Element	Theory	Experiment
Fe	101.79	106[a)]
Co	96.25	110[b)]
Ni	99.04	118[a)]

a) Cited from Ref.9. b) Cited from Ref.10

Table II. Spin-polarized positron lifetimes of bulk Fe (bcc), Co (hcp), and Ni (fcc). We also show the spin moments ($\mu B/atom$).

Element	Spin moment (μB/atom)			Lifetime (ps)		
	Majority spin	Minority spin	Total magnetic moment	$\tau^{\uparrow}$	$\tau^{\downarrow}$	$\tau^{\downarrow} - \tau^{\uparrow}$
Fe	5.2	2.8	2.3	95.159	107.008	11.849
Co	5.3	3.7	1.6	94.493	98.245	3.752
Ni	5.5	4.5	1.0	101.129	96.764	-4.365

Table III. $P^{3\gamma}$ from three-photon polarization measurement

Element	P_{theory}	$P^{3\gamma}_{expt}$ [a)]
Fe	-0.0273	-0.0053
Co	-0.0091	-
Ni	0.0103	0.0012

a) Cited from Ref.4

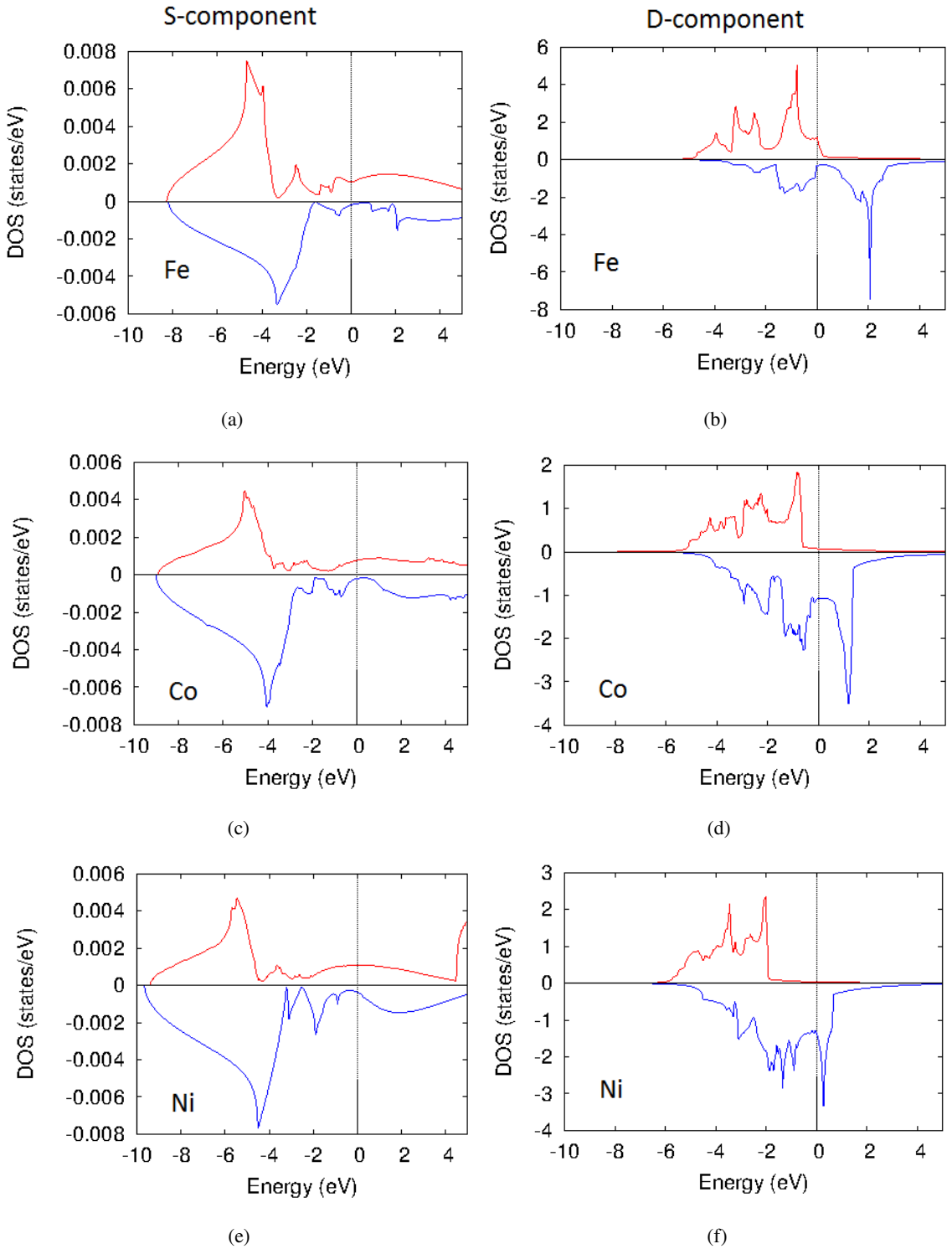

Fig. 1. Atomic projected density of states.

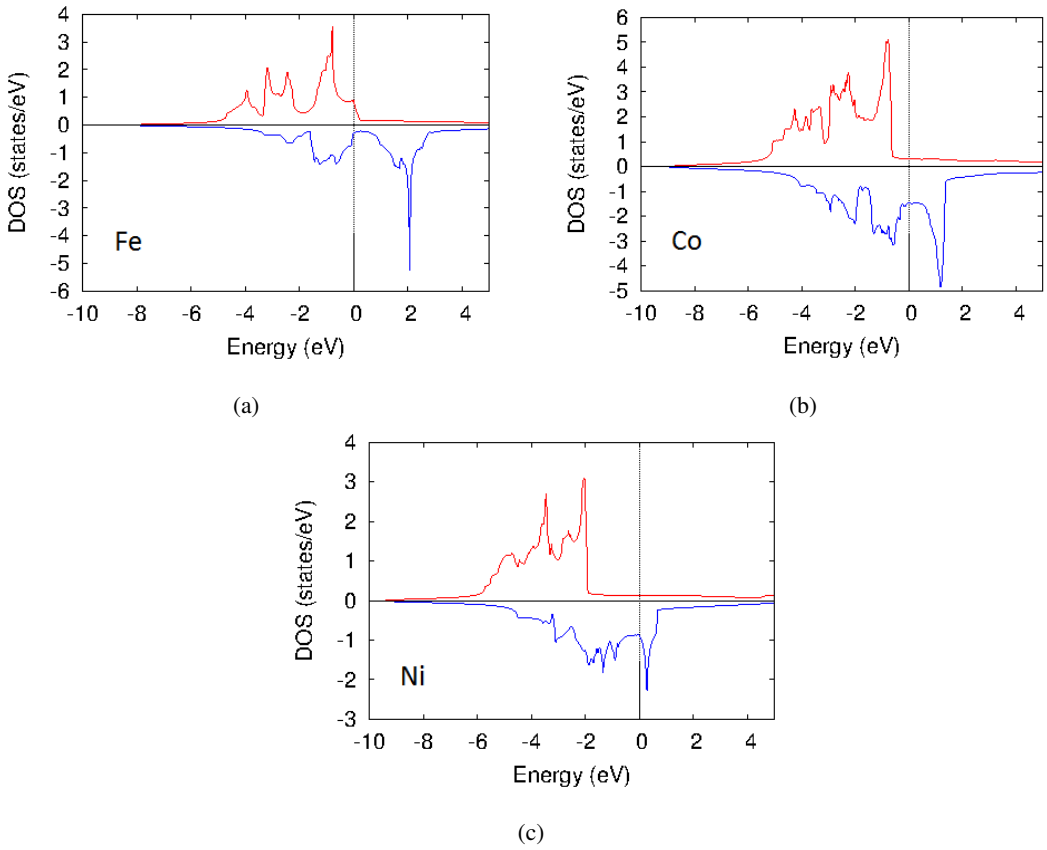

Fig. 2. Spin density of states: (a) Fe, (b) Co, and (c) Ni.

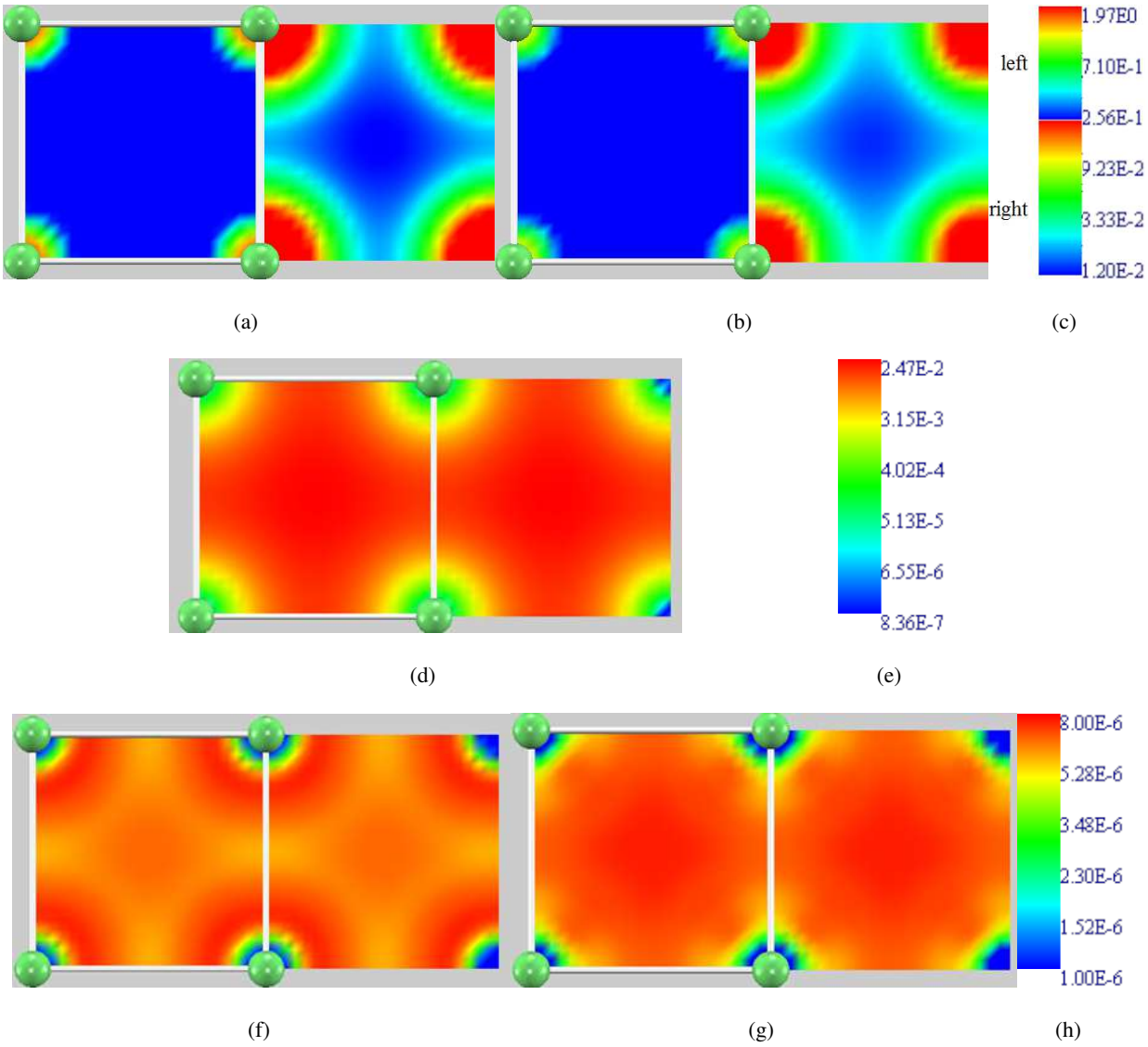

Fig. 3. Majority spin (a) and minority spin (b) densities of electron, density of the positron (d), and electron-positron overlaps for the majority spin (f) and the minority spin (g) in the case of Ni. The units in (a), (b), and (d) are $e/(au)^3$, and those in (f) and (g) are $e^2/(au)^6$.

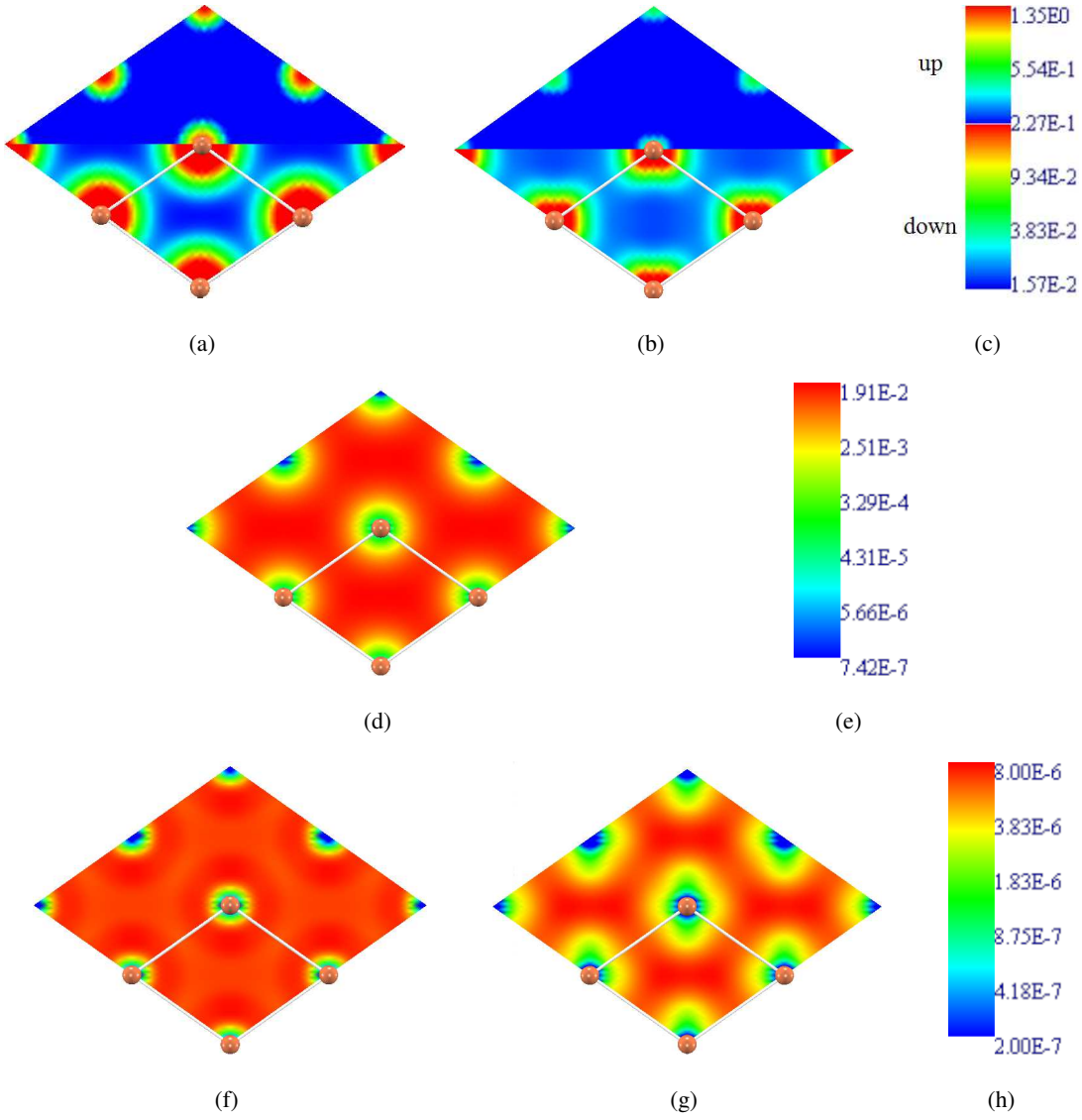

Fig. 4. Majority spin (a) and minority spin (b) densities of electron, density of the positron (d), and electron-positron overlaps for the majority spin (f) and the minority spin (g) in the case of Fe. The units in (a), (b), and (d) are $e/(au)^3$, and those in (f) and (g) are $e^2/(au)^6$.

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