Lifetimes and inelastic mean free path of low-energy excited electrons in Fe, Ni, Pt, and Au: *Ab initio* GW+T calculations

V. P. Zhukov,[1,2] E. V. Chulkov,[1,2] and P. M. Echenique[1,2]
[1]*Departamento de Física de Materiales and Centro Mixto CSIC-UPV/EHU, Facultad de Ciencias Químicas, UPV/EHU, Apdo 1072, 20080 San Sebastián/Donostia, Basque Country, Spain*
[2]*Donostia International Physics Center (DIPC), 20018 San Sebastián/Donostia, Basque Country, Spain*
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We present the results of the GW+T *ab initio* calculations for the inelastic lifetimes and mean free path (IMFP) of low-energy excited electrons in Fe, Ni, Pt, and Au. For Fe and Ni we show that the *T*-matrix terms, incorporating the Stoner's excitations and spin-wave emission, contribute to the IMFP of the spin-minority electrons with excitation energy below 1.2 eV, whereas the GW term dominates at higher energy. We find that the spin dependence of IMFP in Ni is governed mainly by the spin dependence of lifetimes, but in Fe it relates mainly to the spin dependence of group velocities. We show that the "random *k*" model of the electron decay with fixed matrix element well agrees with the GW term of the lifetimes thus showing that the energy and spin dependence of the lifetimes are determined mainly by the convolutions of densities of states. For Au the inclusion of *T*-matrix terms with electron-hole scattering greatly reduces the calculated IMFP, bringing them into better agreement with experimental data.

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I. INTRODUCTION

The transport of excited electrons in ferromagnetic and noble metals is an essential aspect of the physics of magnetoelectronic devices. Among the techniques used to study this phenomenon are spin-resolved electron spectroscopy,[1–3] electron transmission,[4–6] photoemission,[7] time- and spin-resolved two-photon photoemission (TR-2PPE),[8,9] ballistic electron emission microscopy (BEEM).[10–12] Recently ballistic hole magnetic microscopy has been also derived that permits us to study the transport of excited holes in magnetic materials.[13] A characteristic crucial for creating new devices is the attenuation length of hot electrons at low excitation energy. In the interval 0.8–2 eV it can be studied by the techniques of spin-valve transistor (SVT), Ref. 14, and magnetic tunnel transistor (MTT), Refs. 14–17. It has been shown in many experiments that in ferromagnetics the attenuation length of spin-minority hot electrons is much shorter than that of spin-majority electrons.[1–3,5,14] This spin asymmetry is just a phenomenon which makes possible the triggering of current in SVT and MTT devices depending on the direction of external magnetic field. In the majority of papers the spin asymmetry was attributed to spin-dependent lifetimes and interpreted as a consequence of the exchange splitting of energy bands, since this splitting provides the number of unoccupied states for scattering of spin-minority electrons (phase space) larger than that for the scattering of spin-majority electrons. The referenced works contain, however, some essential contradictions. The attenuation length of excited spin-majority electrons in $Ni_{80}Fe_{20}$ measured by MTT (Ref. 16) is almost twice longer than the length from SVT measurements.[14] Both SVT and MTT measurements show that the majority-to-minority ratio of attenuation lengths (4–5) is much larger than the ratio of lifetimes (1.5–2) as measured in TR-2PPE measurements.[8]

It has been shown that the processes of inelastic electron-electron scattering are the most important for the attenuation length.[12,14,16] In Refs. 18 and 19 a phenomenological model of spin-dependent electron transport in SVT has been presented. The model, developed for the attenuation length, incorporates both inelastic and elastic scattering, and the value of inelastic mean free path (IMFP) of excited electrons is one of its parameters. So the knowledge of IMFP is fundamental for the understanding of physical processes in SVT and MTT devices, and the first-principle methods for the calculations of IMFP could be of great help. Previously the first-principle evaluations of IMFP's were performed for nonmagnetic, first of all for free-electron-like metals, see Ref. 20 and references therein. As far as ferromagnetics are concerned, only in the paper of Drouhin[21] the IMFP have been evaluated from a simple model for Fe, Co, and Ni at the excitation energy above 5 eV, but the rest of the papers were dedicated only to studies of lifetimes.

Usually the inelastic electron scattering lifetimes in ferromagnetics are evaluated within various semiempirical versions of scattering theory.[9,21,22] In the most detailed study of Knorren *et al.*[9] the lifetimes of excited electrons in Fe, Co, Ni have been calculated based on the Boltzmann equation and scattering theory with the density of states calculated from first principles. In Refs. 21 and 22 model densities of states and four different transition matrix elements have been employed. It appeared to be possible to adjust the calculated spin asymmetry of lifetimes $\tau_\uparrow/\tau_\downarrow$ to experimentally derived lifetimes. Such evaluations incorporate only a single scattering and do not include the emission of spin waves. Hong and Mills[23] have shown that this emission can be an effective channel for the excited electron decay. A problem of the calculations[23] is that they yield a big contribution of the spin-wave generation to the decay rate of an electron at the excitation energies up to 3 eV, whereas the spin waves are detected in experiments up to the energy below 0.3 eV, see, e.g., Ref. 24.

Recently the lifetimes of excited electrons in Fe and Ni have been calculated from first principles within GW+T

many-body approach.[25] In this approach the lifetimes are evaluated from the imaginary part of the self-energy which is expressed in a series of terms with the dynamically screened potential as a perturbation. The lowest GW-term of the self-energy includes interactions between the excited electron in its initial and all final states by means of the dynamically screened potential. The higher terms of the self-energy are calculated within the *T*-matrix approach which incorporates the creation of secondary electron-hole pairs with subsequent multiple interaction between the primary excited electron and the secondary electrons and holes. An advantage of this method is that it is free of adjusted parameters. Hence it offers a possibility of checking previous models and gaining a more complete insight into the mechanisms of excited electron scattering. In our work, basing on the previous GW+T lifetimes calculations,[25] we evaluate the IMFP of excited electrons in Fe and Ni and compare them with available experimental data for the $Ni_{80}Fe_{20}$ alloy. We also calculate the IMFP in Pt and Au, which are usually employed in the SVT and MTT devices. We examine the role of group velocities and lifetimes in the IMFP and analyze the role of multiple electron-hole scattering in the decay of excited electrons. Additionally we discuss the correctness of two previous concepts, widely used in experimental works and semiempirical evaluations of lifetimes, namely, the concept of "phase space" and the "random *k*" approximation.

II. METHOD OF CALCULATION

The mean free path of an excited electron in a one-electron state $\psi_{\mathbf{q}n\sigma}$ with momentum $\mathbf{q}$, band index n, spin σ, energy $\epsilon_{\mathbf{q}n\sigma}$, and group velocity $\mathbf{v}_{\mathbf{q}n\sigma}$ is the distance the electron travels during its lifetime $\tau_{\mathbf{q}n\sigma}$, $\lambda_{\mathbf{q}n\sigma}=v_{\mathbf{q}n\sigma}\tau_{\mathbf{q}n\sigma}$. The group velocity is defined as $\mathbf{v}_{\mathbf{q}n\sigma}=\partial\epsilon_{\mathbf{q}n\sigma}/\partial\mathbf{k}$.[26] Modern *ab initio* methods of lifetimes calculations are based on the self-energy formalism of many-body theory.[27] In the calculations for real materials the GW approach (GWA) is usually employed, where the electron self-energy Σ is calculated retaining only the first term in the series expansion of Σ in terms of the spin-independent screened Coulomb potential W

$$\Sigma_\sigma(1,2)=iG_\sigma(1,2)W(1,2). \tag{1}$$

Here G_σ is the Green's function of the excited electron with spin σ, and $1\equiv(\mathbf{r}_1,t_1)$. It is well known that the GWA is fairly good for systems with dominating long-range screening, but it is not accurate enough to describe short-range interaction. The self-energy of an electron with spin σ is determined in the GWA by the Green's function with the same spin, so GWA does not include spin-flip processes which can be important in spin-polarized materials. In order to avoid the limitations of the GWA, in Refs. 25 and 28 an *ab initio* GW+T method of self-energy calculations has been proposed to account for the most important high-order scattering processes by means of the *T*-matrix technique.[27] The *T*-matrix operator is defined as the solution of the Bethe-Salpeter equation[27]

$$T_{\sigma_1,\sigma_2}(1,2|3,4)=W(1,2)\delta(1-3)\delta(2-4)+W(1,2)\int d1'd2' \times K_{\sigma_1,\sigma_2}(1,2|1',2')T_{\sigma_1,\sigma_2}(1',2'|3,4). \tag{2}$$

Depending on the form of the kernel K_{σ_1,σ_2}, the T matrix describes different multiple scattering processes. In transition metals at low excitation energy the most important is the direct electron-hole scattering, when kernel K is a product of particle and hole Green's functions

$$K_{\sigma_1,\sigma_2}(1,2|1',2')=iG^h_{\sigma_1}(1,1')G^e_{\sigma_2}(2',2). \tag{3}$$

The sum of direct self-energy terms is expressed as

$$\Sigma^d_{\sigma_2}(4,2)=-i\sum_{\sigma_1}\int d1d3G^e_{\sigma_1}(3,1)T_{\sigma_2,\sigma_1}(1,2|3,4). \tag{4}$$

So, in addition to GWA, the *T*-matrix approach includes multiple non-spin-flip and spin-flip scattering processes. For ferromagnetics it means that the scattering of excited electrons as accompanied by the creation of spin waves and Stoner's electron-hole pairs is taken into account.

The details of the GW+T approach can be found in Refs. 25, 28, and 29. The calculations of the lifetimes of excited electrons in Fe and Ni (Ref. 25) have shown that at the excitation energy below 0.5 eV the *T*-matrix self-energy term is important for spin-minority electrons in Fe. It will be shown below that for the excitation energies above 1.2 eV the IMFP of electrons in Fe and Ni is mainly defined by the scattering processes included in the GWA, whereas the contribution of the T matrix is insignificant. Therefore, here we discuss mainly the GWA calculations, although we will see that the *T*-matrix electron-hole scattering is important in Au too.

Within the GWA, the lifetime of an electron in the state $\psi_{\mathbf{q}n\sigma}$ is governed by the expectation value of the imaginary part of self-energy $\mathrm{Im}\,\Sigma_{\mathbf{q}n\sigma}(\omega)\equiv\langle\psi_{\mathbf{q}n\sigma}|\mathrm{Im}\,\Sigma_\sigma(\omega)|\psi_{\mathbf{q}n\sigma}\rangle$. We perform the GWA calculations by expanding the self-energy and all related values in the basis of Bloch functions $B_{\mathbf{k}i}$ constructed from the products of linear muffin-tin orbitals (LMTO).[30,31] So we have for excited electrons ($\omega>E_F$)

$$\mathrm{Im}\,\Sigma_{\mathbf{q}n}(\omega)=-\sum_{\mathbf{k}}^{\mathrm{unocc}}\sum_{n'}\sum_{i,j}\mathrm{Im}\,W_{i,j}(\mathbf{k},\omega-\epsilon_{\mathbf{k}-\mathbf{q}n'\sigma}) \times\langle\psi_{\mathbf{q}n\sigma}\psi_{\mathbf{k}-\mathbf{q}n'\sigma}|B_{\mathbf{k}i}\rangle\langle B_{\mathbf{k}j}|\psi_{\mathbf{k}-\mathbf{q}n'\sigma}\psi_{\mathbf{q}n\sigma}\rangle \times\Theta(\omega-\epsilon_{\mathbf{k}-\mathbf{q}n'\sigma}). \tag{5}$$

The key value in the evaluations of $\mathrm{Im}\,W(\omega)$ is the polarization function $\mathbf{P}(\omega)$ whose calculations with the basis functions $B_{\mathbf{k}j}$ have been discussed in details in Ref. 30. Once the polarization matrix is obtained, we evaluate the matrix of the density-density response function $\mathbf{R}$, dielectric and inverse dielectric matrices ϵ and ϵ^{-1}, and then calculate the matrix of screened Coulomb interaction $\mathbf{W}$:

$$\mathbf{R}(\omega)=[\mathbf{1}-\mathbf{V}\cdot\mathbf{P}(\omega)]^{-1}\mathbf{P}(\omega), \tag{6}$$

$$\epsilon(\omega)=\mathbf{1}-\mathbf{V}\cdot\mathbf{P}(\omega), \tag{7}$$

$$\epsilon^{-1}(\omega) = \mathbf{1} + \mathbf{V} \cdot \mathbf{R}(\omega), \tag{8}$$

$$\mathbf{W}(\omega) = \epsilon^{-1}(\omega)\mathbf{V}. \tag{9}$$

Then $\mathrm{Im}\,\Sigma(\omega)$ can be evaluated, and the real part of self-energy $\mathrm{Re}\,\Sigma(\omega)$ can be obtained from the Hilbert transform of $\mathrm{Im}\,\Sigma(\omega)$, see Ref. 30.

The many-body self-energy corrections to the eigenvalues $\epsilon_{\mathbf{q}n\sigma}$ are determined by the expectation values of the operator $\Delta\Sigma_\sigma(\omega) = \Sigma_\sigma(\omega) - V_\sigma^{\mathrm{xc}}$ where V_σ^{xc} is the exchange-correlation potential. Namely, we have the Dyson equation for the complex electron energy

$$E_{\mathbf{q}n\sigma}(\omega) = \epsilon_{\mathbf{q}n\sigma} + \langle \psi_{\mathbf{q}n\sigma} | \Delta\Sigma_\sigma(\omega) | \psi_{\mathbf{q}n\sigma} \rangle. \tag{10}$$

There are two approximate methods of solving Eq. (10). First is a so-called "on shell approximation" when it is approved that in Eq. (10) $\omega = \epsilon_{\mathbf{q}n\sigma}$. The second, presumably more exact approach is when the equation is solved by retaining linear part of the dependence of $\mathrm{Re}\,\Delta\Sigma_\sigma$ on ω near the eigenvalue $\epsilon_{\mathbf{q}n\sigma}$ and neglecting the variation of $\mathrm{Im}\,\Delta\Sigma_\sigma$. In this approximation the self-energy corrections to the LDA eigenvalues are

$$\Delta\epsilon_{\mathbf{q}n\sigma} = E_{\mathbf{q}n\sigma} - \epsilon_{\mathbf{q}n\sigma} = Z_{\mathbf{q}n\sigma}\Delta\Sigma_{\mathbf{q}n\sigma}(\omega = \epsilon_{\mathbf{q}n\sigma}), \tag{11}$$

where

$$Z_{\mathbf{q}n\sigma} = \left[1 - \frac{\partial\, \mathrm{Re}\,\Delta\Sigma_{\mathbf{q}n\sigma}(\omega)}{\partial\omega} \right]^{-1}_{\omega=\epsilon_{\mathbf{q}n\sigma}} \tag{12}$$

is the renormalization factor. This way is reduced to the "on shell" approximation if $Z=1$. The imaginary part of the self-energy correction gives then the line-width of the excitation, and the inverse value determines the lifetime of excitation[27]

$$\tau^{-1}_{\mathbf{q}n\sigma} = 2Z_{\mathbf{q}n\sigma} |\mathrm{Im}\,\Delta\Sigma_{\mathbf{q}n\sigma}|. \tag{13}$$

Having calculated Z, one can also evaluate velocities corrected for the self-energy effects

$$v_{\mathbf{q}n\sigma} = \partial\epsilon_{\mathbf{q}n\sigma}/\partial\mathbf{k} + \partial\,\mathrm{Re}\,\Delta\Sigma_{\mathbf{q}n\sigma}/\partial\mathbf{k} = v_{\mathbf{q}n\sigma}[2 - 1/Z_{\mathbf{q}n\sigma}]. \tag{14}$$

Typical Z values for transition metals are within 0.6–0.8, so in the calculations of IMFP the Z value of the Eq. (13) almost compensates the Z dependent brackets in Eq. (14). Therefore we do not find any advantage of the IMFP evaluated with exact Z over the IMFP with $Z=1$ applied both for τ and v—see discussion below.

A valuable property of the GWA within the "on shell" approximation is that it allows one to interpret the first-principle lifetimes in terms of a transparent and easily calculated band-structure characteristic, a double convolution of density of state. It follows from Eqs. (5)–(9) that

$$\mathrm{Im}\,\mathbf{W} = \mathbf{V}\,\mathrm{Im}[(\mathbf{1} - \mathbf{P}\cdot\mathbf{V})^{-1}]\mathbf{V}. \tag{15}$$

The poles of the function $(\mathbf{1} - \mathbf{P}\cdot\mathbf{V})^{-1}$ define the energy of plasmons which is in transition metals typically higher than the excitation energy under consideration. Neglecting these poles, that is considering only the deexcitation accompanied by creation of electron-hole pairs, we have

$$\mathrm{Im}\,\mathbf{W} \simeq \mathbf{V}\,\mathrm{Im}\,\mathbf{P}\cdot\mathbf{V}. \tag{16}$$

We also apply to the Eq. (5) the approximation of a unique frequency independent transition matrix element M^2 which incorporates both the matrix elements $\mathrm{Im}\,W_{i,j}$ and the integrals $\langle\psi\psi|B\rangle$ of Eq. (5). Normally this is called a "random k" approximation.[33] Finally, at zero temperature we come to the expression

$$\mathrm{Im}\,\Sigma_\sigma(\epsilon) = \int_{E_F}^{\epsilon} d\omega\rho_\sigma(\omega) \int_{E_F-(\epsilon-\omega)}^{E_F} d\omega'[\rho_\sigma(\omega')\rho_\sigma(\epsilon-\omega-\omega') + \rho_{-\sigma}(\omega')\rho_{-\sigma}(\epsilon-\omega+\omega')] \times M^2, \tag{17}$$

where ρ_σ is the spin-resolved density of states. The external integral of this double-convolution counts the number of states available for the deexcitation of a primary excited electron with energy ϵ (phase space). The internal integral is a convolution of density of states that counts all the electron-hole pairs created at the score of the energy lost by the primary electron.

Equation (17) is similar to the equations for the decay rate that follow from the "golden rule" of scattering theory. Such equations were widely used before in semiempirical calculations for lifetimes and bandwidths, see Refs. 9, 21, 22, and 32. More complicated expressions were also employed in these papers which included up to four different matrix elements. We will show below that at the energy of excitation up to 3 eV even with one matrix element the expression (17) well describes the results of the GWA calculations.

Normally, the IMFP's deduced from experiments with ferromagnetics are not resolved in momentum, but resolved in spin; in Ref. 16 the energy dependencies of IMFP's are also given. Therefore we calculated momentum-averaged and energy-resolved values of velocities $v_\sigma(E)$, lifetimes $\tau_\sigma(E)$, and IMFP's $\lambda_\sigma(E) = v_\sigma(E)\tau_\sigma(E)$.

We evaluated the velocities from the band-structure calculations by means of the LMTO method, Ref. 34. As far as experiments had been performed with polycrystalline films, we averaged the velocities calculated for the radial directions in the Brillouin zone. The velocities strongly depend on the number of momentum vectors $\mathbf{k}$ in the Brillouin zone (BZ). A good averaging of velocities is achieved with about 5000 $\mathbf{k}$ in the irreducible BZ. Contrary to the calculations of velocities, the momentum averaging of lifetimes is well converged at a rather small number of $\mathbf{k}$ vectors, about 300 in the irreducible BZ.

A second possible way of calculating momentum-averaged IMFP's $\lambda(E)$ is to perform averaging of momentum-resolved IMFP's $\lambda_{\mathbf{q}n\sigma} = v_{\mathbf{q}n\sigma}\tau_{\mathbf{q}n\sigma}$. We revealed that this kind of averaging yields almost the same results as the first way of separate averaging for velocities and lifetimes, so we discuss below only the IMFP's obtained in the first way.

III. RESULTS AND DISCUSSIONS

In Figs. 1 and 2 we show our GWA IMFP's for Ni and Fe. They are given for the "calculated Z" and "on shell" approximations. For spin-minority electrons we give also the results of the GW+T calculations. Since the experimental data for

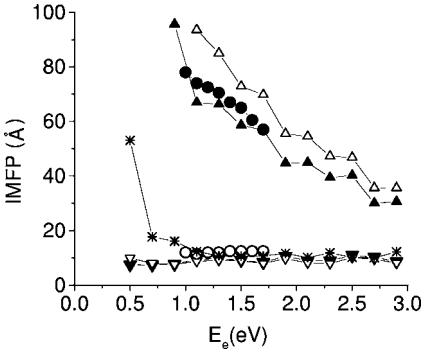

FIG. 1. The calculated IMFP's in Ni with respect to excitation energy E_e. Solid triangles are results with calculated Z; open triangles are results with $Z=1$. Up triangles show spin-majority GWA IMFP's. Down triangles show spin-minority GWA IMFP's corrected for T-matrix effects for both spin-flip and non-spin-flip processes. Stars show the GWA results for spin-minority electrons without T-matrix corrections. Solid (open) circles are experimental data for spin-majority (−minority) electrons in $Ni_{80}Fe_{20}$ from Ref. 16.

pure Ni and Fe are absent, we compare the calculated IMFP's with the energy-resolved experimental attenuation length's for $Ni_{80}Fe_{20}$ from Ref. 16.

We find that both in Ni and Fe the T-matrix corrections to spin-minority IMFP's are comparable with the GW term at small energy, but at the energy above 1.2 eV the T-matrix corrections are negligible. Hence the most important process of an excited electron decay is this included in the GWA theory, i.e., a single scattering of an excited electron with transfer of energy to a virtual non-spin-flip electron-hole pair.

Comparing the GWA data on IMFP obtained within the "calculated Z" and the "on shell" approximations, we find that the difference between them is rather insignificant: it is negligible for the IMFP of spin-minority electrons, and is only about 20% for the IMFP of spin-majority electrons. Both calculations correspond equally well to the trends in the experimental data for $Ni_{80}Fe_{20}$. These trends are a rapid decrease with energy for the big spin-majority IMFP and the invariance for the small spin-minority IMFP.

So we do not find that the presumably more exact "calculated Z" approach yields the results better than the "on shell"

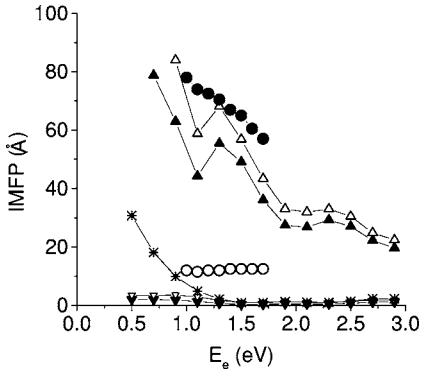

FIG. 2. The calculated IMFP's in Fe and experimental IMFP's in $Ni_{80}Fe_{20}$. Notations are as in Fig. 1.

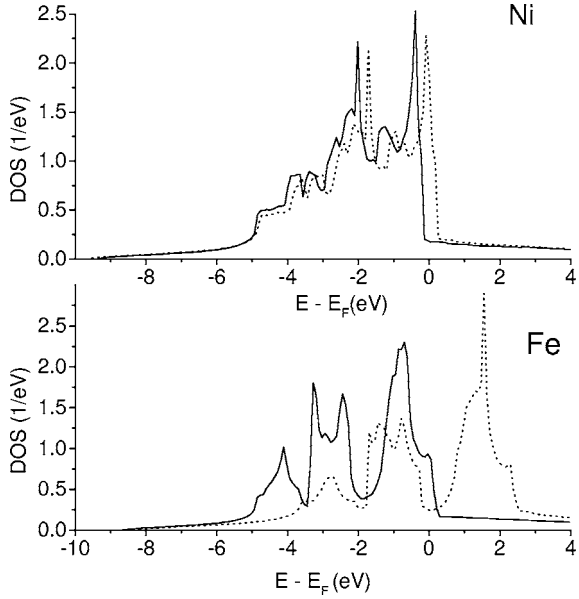

FIG. 3. The densities of states in Fe and Ni. Solid lines are for spin-majority electrons whereas dotted lines are for spin-minority electrons. The energy of states is given with respect to the Fermi level E_F.

approach, hence we discuss below the results of the simpler "on shell" calculations. We see that in Ni the spin asymmetry of the IMFP, i.e., the ratio of the spin-majority IMFP to the spin-minority IMFP, decreases from ~7 at the energy 1 eV to ~3 at the energy 3 eV. In Fe the spin asymmetry is much higher due to the lower values of the IMFP's for spin-minority electrons. The energy and spin dependencies of the IMFP find explications based on the spin-resolved densities of states in Ni and Fe shown in Fig. 3. In Ni at the energy from 1 to 2 eV both the spin-majority and spin-minority electronic states are free-electron-like, so high velocities are expected for these states. In Fe in this energy region the spin-majority states are free-electron-like whereas the spin-minority states are 3d-like. So the velocities of the Fe spin-majority electrons should be also high, but the velocities of the spin-minority electrons should be lower. This is well confirmed by the data given in Fig. 4. It shows that in Ni the velocities of the spin-majority and spin-minority electrons differ by about 30%, whereas in Fe this difference is much higher. Spin-minority velocity in Fe only slightly varies at the energy below 2.3 eV, but it markedly increases at higher energy, when the electronic states become free-electron-like.

In Fig. 5 we show the electron lifetimes in Ni from the GWA and from the "random k" approximation. The spin asymmetry of lifetimes in Ni appears to be much higher than in Fe, in particular at small energy of excitations. So we conclude that the spin asymmetry of IMFP's in Ni and Fe are governed by different factors: in Ni the main factor is the difference in lifetimes whereas in Fe it is the difference in velocities. It becomes evident also that the IMFP of spin-minority electrons in Fe are very small because of the very small velocities of electrons.

The averaged GWA electron lifetimes in Ni are almost perfectly reproduced in the "random k" approximation, Eq.

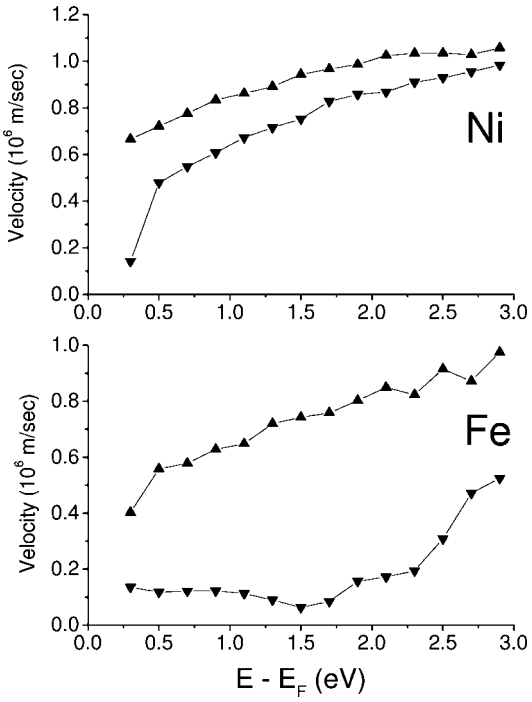

FIG. 4. The momentum-averaged electron velocities in Ni and Fe. Up triangles are for spin-majority electrons and down triangles are for spin-minority electrons.

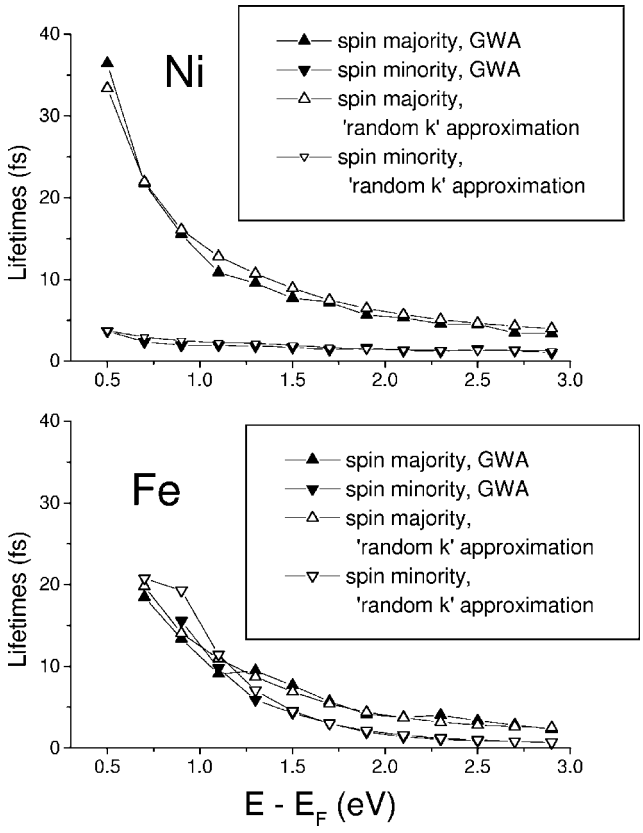

FIG. 5. The momentum-averaged electronic lifetimes in Ni and Fe as calculated in GWA and "random k" approximation.

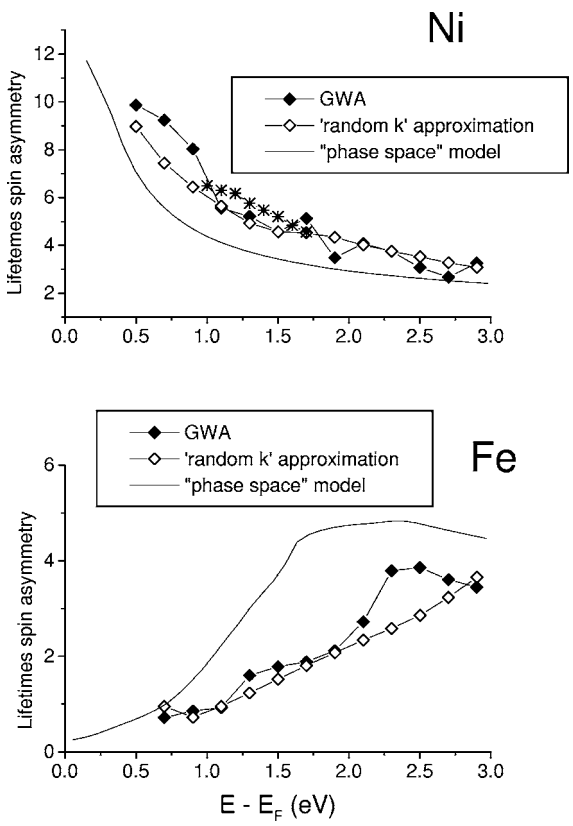

FIG. 6. The spin asymmetry of electronic lifetimes in Ni and Fe as calculated within GWA, "random k" approximation and "phase space" model. In the upper panel the stars show also the experimental spin asymmetry of attenuation lengths in the alloy $Ni_{80}Fe_{20}$, Ref. 16.

(17), with the value of matrix elements $M^2=0.075$ eV^2 equal for both spins. With the same value of the matrix element, the "random k" lifetimes of the spin-majority free-electron-like electrons in Fe also well agree with the GWA lifetimes, whereas for 3d-like spin-minority states a good agreement is achieved at $M^2=0.06$ eV^2. So we find that the energy and momentum dependencies of the matrix elements of GWA are irrelevant for averaged lifetimes, and the energy and spin dependence of lifetimes is governed mainly by the convolution of densities of states, as described by Eq. (17). This conclusion is in accord with the previous calculations showing that the energy dependencies of averaged lifetimes in Nb, Mo, Rh, Pa, and Ag also are well reproduced in the "random k" model.[35]

In Fig. 6 we show the spin asymmetry of electronic lifetimes in Ni and Fe as calculated within the GW approach, "random k" approximation and "phase space" model. We see that in Ni the GWA spin asymmetry is typically higher than the asymmetry in Fe, and the asymmetries in Ni and Fe have different trends with energy, i.e., decrease with energy in Ni and increase in Fe. The "random k" approximation (which in the calculations of asymmetry has no adjustable parameters) well reproduces the GWA results both for Fe and Ni, apart from some relatively small deviations. Although the results of the "phase space" model are markedly worse, they qualitatively correspond to the trends, which help us to under-

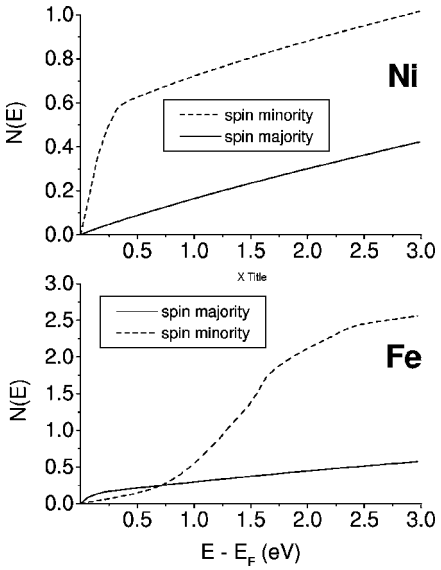

FIG. 7. The "phase spaces" in Ni and Fe with respect to the excitation energy $E-E_F$.

stand their origin. In the "phase space" model the spin asymmetry is evaluated as $\tau_\uparrow(E)/\tau_\downarrow(E)=N(\downarrow)(E)/N(\uparrow)(E)$ where phase space $N(E)$ is the number of states between the Fermi energy and the energy of excited state E. In Fig. 7 we show the energy dependencies of the phase spaces in Ni and Fe. Comparing with Fig. 3, for Ni we see that at the excitation energy $E_e=E-E_F\leqslant 0.2$ eV the spin-minority phase space is much bigger than the spin-majority phase space due to the presence of the spin-minority d states, hence defining big values of spin asymmetry. Above 0.2 eV both spin-majority and spin-minority states are free-electron-like with small density of states. So with further increase of the excitation energy the spin asymmetry in Ni slowly converges to 1. In contrast to Ni, in Fe the spin-majority states dominate near the Fermi level, whereas the density of spin-minority states is at minimum. This defines the small value of asymmetry (<1) at small excitation energy. With the increase of excitation energy the phase space of the spin-minority electrons greatly increases due to the presence of the broad and high band with maximum around 1.3 eV whereas the phase space of the free-electron-like majority states changes slowly. This defines the increase of the spin asymmetry in Fe.

The GWA and "random k" IMFP's in Ni appear to be in agreement with the experimental attenuation lengths for the alloy $Ni_{80}Fe_{20}$ from Ref. 16. It is confirmed also that the spin asymmetry of IMFP's is governed for this alloy mainly by the effects of lifetimes.[14] We see, however, that the spin asymmetry of the GWA lifetimes in Ni, about 7 at 1 eV, is much higher that the spin asymmetry of relaxation times as measured in the TR-2PPE experiments,[9] about 1.5 at 1 eV. In addition, the TR-2PPE spin asymmetry is much less than the spin-asymmetry of attenuation lengths from the SVT measurements.[14] The origin of these disagreements is not clear; probably, in the experiments[9] an important role is played by the factors discarded in the GWA approximation, such as scattering of excited electrons with phonons and impurities, effects of cascade electrons and transport. So new

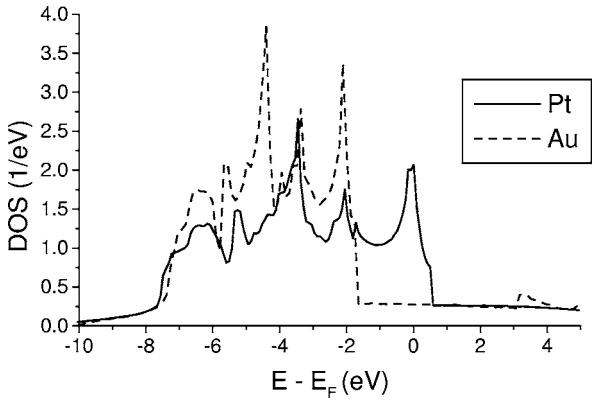

FIG. 8. The total densities of states in Pt and Au.

efforts, with an analysis of all the given mechanisms of electron scattering, are desirable for Ni, Fe, Co.

Our calculations help to estimate the choice of IMFP's in the Ref. 19, where the IMFP's were input parameters in the model of transport effects in spin-valve transistor. In Ref. 19 the IMFP's of spin-majority and spin-minority electrons in $Ni_{80}Fe_{20}$ were evaluated at 1 eV as equal to 160 and 80 Å, respectively. The value of electron velocity has been chosen in Ref. 19 rather arbitrarily as equal to 2×10^6 m/sec. This velocity is much higher than our first-principle velocities, which makes clear why the IMFP's of Ref. 19 are about twice as big as our results. It was also adopted in Ref. 19 that the lifetimes in $Ni_{80}Fe_{20}$ are equal to the relaxation times in Co from the TR-2PPE measurements,[9] with the spin-asymmetry about 2 at excitation energy 1 eV. However, such value of spin asymmetry is much less than the spin asymmetry of lifetimes from our calculations or spin-asymmetry of attenuation lengths from the SVT measurements.[14]

In Fig. 8 we show the densities of states in Pt and Au. The states in Pt with the energy from -10 eV up to 0.7 eV are d-like states, whereas the states above are free-electron-like. The DOS of Pt is very similar to that of Pd, whose lifetimes we calculated in the GW+T approach in Ref. 29. The calculations for Pd had shown a rather small contribution of the T-matrix term which was 20–30 % of the GWA term, so for Pt we calculated only the GWA term. The top energy of d states in Au is -1.8 eV, hence, contrary to Pt and Pd, the lifetimes are determined mainly by the free-electron-like states. This case is similar to the case of Al where the T-matrix terms are essential for lifetimes.[29] Therefore, together with the GWA term, we calculated for Au also the T-matrix terms with multiple electron-hole and electron-electron scattering. The electron-electron T-matrix term appeared to be much less than the electron-hole term, so in the following we omit this term.

In Fig. 9 we show velocities and lifetimes in Pt and Au, and in Fig. 10 the corresponding IMFP's. Since in Pt the states below 0.7 eV are d-electron-like, their velocities are much less than the velocities of the electrons in Au. At higher energy the velocities of the electrons in Pt and Au differ only by about 20–30 %, which is about 10 times less than the difference in IMFP's in Fig. 10. So the difference of the IMFP's in Pt and Au is governed mainly by the lifetimes.

The electrons in Au have much longer lifetimes than the electrons in Pt. Smaller lifetimes in Pt are explained by the

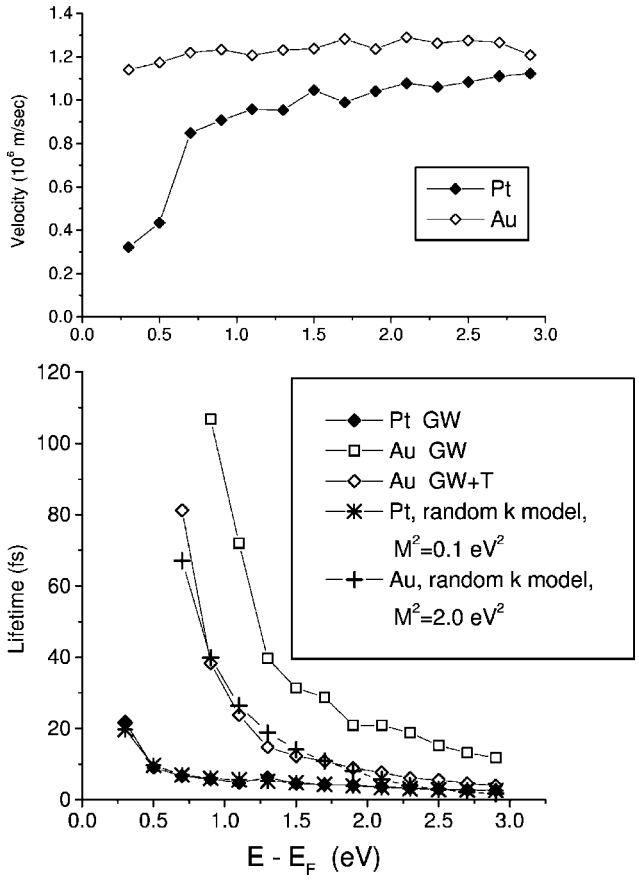

FIG. 9. The velocities and lifetimes of electrons in Pt and Au.

presence of a high peak of the DOS near the Fermi level. This peak essentially increases both the phase space of the primary electron decay, that is the external integral in Eq. (17), and the phase space of the polarization, that is the internal integral in the same equation. The electrons in Au have smooth and small density of states, so the lifetimes are very long. The energy change of lifetimes in Pt is well reproduced in the "random k" approximation with energy-independent matrix element M^2. This is valid also for the lifetimes in Au as calculated by means of the GW+T method.

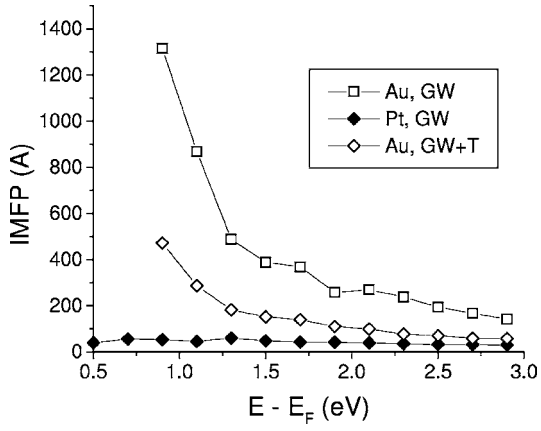

FIG. 10. The inelastic mean free paths of electrons in Pt and Au.

We find that in Au the T-matrix term with multiple electron-hole scattering reduces the lifetimes by a factor of 2. An analogous result was found before for Al where the lifetimes calculated without T-matrix term are much higher than the experimental data.[29] We refer the effect of T-matrix in these metals to a rather high value of the screened potential, due to which the processes of multiple scattering become essential. At the excitation energy about 1 eV we have the GWA IMFP in Au equal to 1300 Å. This is unreasonably higher than the attenuation length deduced from the photoemission measurements, 220–230 Å, see Ref. 36, and from BEEM measurements, 230–280 Å, see Ref. 11. Our GW+T IMFP in Au at 1 eV is equal to 470 Å, so we see that the inclusion of the T-matrix terms brings the calculated IMFP in better agreement with experimental data. The value of IMFP in Au, proposed in Ref. 19, is equal to 400 Å, i.e., in good agreement with our results.

To our knowledge, experiments have not been done on the IMFP's in Pt. It was supposed in Ref. 19 that the attenuation length in Pt might be close to that in PtSi, 40 Å, and based on this supposition the IMFPs in Pt have been evaluated as equal to 100 Å. This is much higher than our IMFP value, 50 Å. However, the evaluations in Ref. 19 are in disagreement with the data of Ref. 37 where it has been shown, both from photoemission measurements and first-principle calculations, that the band states in PtSi above the Fermi level are mostly states of Si and not the states of Pt. So the evaluation of the attenuation length and IMFP in Pt, as an input parameter in the model of transport, probably, needs a revision.

IV. CONCLUSIONS

In this paper we have reported on the first-principles calculations of inelastic lifetimes and mean free paths of excited electrons in metals commonly used in spintronic devices: Ni, Fe, Pt, and Au. The lifetimes were evaluated from the self-energy of the excited electrons by means of the first-principle GW+T approach. Our calculations for the self-energy included, together with the traditional GWA term, also higher terms of the many-body perturbation theory which describe multiple electron-hole scattering within formalism of the T-matrix. For ferromagnetics, the T-matrix theory incorporates the processes of deexcitations through the creation of Stoner's pairs and spin waves, and also non-spin-flip multiple scattering processes.

We found that in the calculations of IMFP in Ni and Fe the T-matrix term is essential for the excitation energies less than 1.2 eV, but for higher energies the GWA term definitely dominates. The GWA calculations yield the results that well agree with the experimental attenuation lengths for the alloy $Ni_{80}Fe_{20}$. The calculations well reproduce the variation of the experimental spin-resolved IMFP's and spin asymmetry of the IMFP's with the energy of excitation. We show that the spin dependence of lifetimes is the main factor that determines the spin asymmetry of IMFP's in Ni whereas in Fe the spin dependence of velocity is more important.

We show that neglecting the participation of plasmons in the decay of electron excitations, one can reduce the GWA expression for the imaginary part of self-energy to the so-

called "random k" approximation. In this approximation the decay rate is evaluated from an easily calculated and physically transparent double convolution of density of states scaled with a unique momentum- and energy-independent matrix element. We demonstrate that the "random k" approximation, with the double convolution calculated from the first-principles densities of states, well corresponds to the energy dependence of GWA lifetimes in Fe, Ni, Pt, Au and also to the calculated spin asymmetry of IMFP's in Fe and Ni. For the "phase space" model of decay we find that, although the correspondence between this model and GWA calculations is markedly worse, on a qualitative level the "phase space" model correctly reproduces the energy dependence of lifetimes and spin asymmetry of lifetimes. It makes it possible, based on the "phase space" model, to explain the increase of the spin asymmetry of lifetimes in Fe with energy by the presence of the band of spin-minority states above the Fermi level.

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