

Calculation of low-energy-electron lifetimes

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The effect of the density of states on the lifetime of low-energy electrons in metals has been studied using a golden rule approach. Simple approximations to the real density of states of metals have been used allowing analytical results, which show that the free-electron scaling $(E - E_F)^{-2}$ of the lifetime is affected above the onset of d -electron contributions. Hence, in noble metals a scaling with $(E - E_F - \omega_d)^{-2}$, where ω_d is the energy distance from the Fermi energy to the top of the d band, appears once d electrons can be excited. In ferromagnetic Co, the ratio between the lifetimes of majority and minority spin electrons is found energy independent below the threshold for the excitation of majority d electrons while this ratio increases with energy above that point. [S0163-1829(99)09327-3]

I. INTRODUCTION

The transient behavior of excited electrons at or near metal surfaces is fundamental to many important surface processes, such as chemical reactions, transport, surface-phase transitions, molecule-surface interactions, as well as in technological applications of electronic materials.^{1,2} The development of new experimental techniques such as time-resolved two-photon photoemission (TR-2PPE) allows to measure the lifetime of excited quasiparticle states in metals on a femtosecond scale with high precision.³⁻¹⁰ These kind of experiments, although their interpretation is still not settled, indicate that a description more accurate than the one given by the free-electron approach¹¹ is necessary. Such lifetime calculations have a long story.¹² Quinn¹³ also introduced the effect of a polarizable background with a static dielectric constant ϵ_0 showing that the lifetimes increased as $\sqrt{\epsilon_0}$ since the electron-electron interaction is reduced due to the additional screening. Krolikowsky and Spicer¹⁴ used experimental density of states (DOS) to calculate the qualitative behavior of the lifetime, which was compared to the pioneering work by Kanter¹⁵ who measured lifetimes in Ag, Al, and Au in transmission experiments for electrons about 10 eV above the Fermi energy. There are many studies related to the calculation of the electronic mean-free path (l) both for ions and electrons, whose magnitude is directly related to the lifetime by the velocity (v), $l = v\tau$.

Using the self-energy formalism, Shelton¹⁶ calculated the inelastic mean-free path of electrons in jellium for energies from 10 to 500 eV above the Fermi energy. In the same formalism, Penn¹⁷ introduced the effects of exchange and correlation and calculated the mean-free path for different metals and insulators. Tung, Ashley, and Ritchie¹⁸ used a statistical approach to calculate the mean-free path as an average of the free-electron mean-free paths in each point of the unit cell in the energy range between 1 and 10^5 eV, and

studied the effect of core polarizability, damping, and local-field corrections on the mean-free path in Aluminum between 10 and 50 eV.¹⁹ With a similar approach as in Ref. 18, Penn²⁰ used optical response functions to calculate the mean-free path within a dielectric formalism from 5 up to 10^4 eV. Ashley²¹ proposed a prescription for constructing a dielectric-response function including dispersion effects, from the optical one, and calculated the mean-free path and the energy loss for electrons between 40 and 10^4 eV in noble metals.

In ferromagnets, Penn, Apell, and Girvin²² made a study of the secondary electron cascades, which allows to obtain information about the lifetimes from the polarization of the emitted secondaries. Several experiments in ultrathin magnetic layers have confirmed the spin dependence of the lifetime in these materials (see e.g., Refs. 23-25). Siegman²⁶ showed that d electrons should play an important role in understanding the mean-free path in transition metals. Passek and coworkers²⁷ measured spin-dependent lifetimes of image states in Fe[110] and analyzed the data by calculating the convolution of first-principles DOS. Recently, Drouhin²⁸ developed a detailed mathematical approach to the problem of spin-dependent lifetimes in ferromagnetic materials in terms of the density of states.

In this paper, we try to delineate the effect of the density of states (DOS) on the lifetime of low-energy electrons ($E - E_F < 4$ eV). Usually the approximation of assuming a constant DOS is made but in the case of transition metals, with d bands near or even at the Fermi energy, one expects that such an assumption will not provide a good description. The well-known scaling of the lifetime as $(E - E_F)^{-2}$ is a direct consequence of assuming a smooth DOS in the energy region of the involved electronic transitions, that allows a Taylor expansion around the Fermi energy. This expansion will only be valid very close to the Fermi energy. We have thus found that the mentioned scaling property of the lifetime is

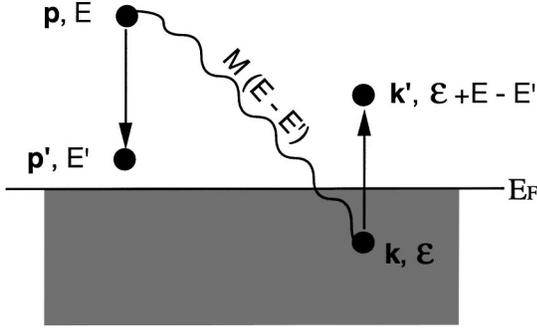


FIG. 1. Scheme for the deexcitation process in a scattering picture.

affected by the inclusion of a more realistic DOS once the energy of the electron is larger than the interband excitation energy.

In order to model the d bands, we have used simple descriptions that allow analytic expressions for the lifetime in the approximation of constant matrix elements. They give the same characteristic features as calculations based on full first-principles DOS. Within this model we are able to describe in a reasonable way the available experimental data.

In the next section, we use our approach to study the behavior of the lifetime in noble metals (Cu and Ag). In Sec. III, we generalize the model to a ferromagnet (Co) in which a spin-dependent lifetime is expected. In Sec. IV, we analyze the available experimental data in terms of our approach. Finally, in Sec. V we discuss the major conclusions from our study.

II. LIFETIME IN NOBLE METALS

Consider an electron with momentum \mathbf{p} and energy E above the Fermi energy. The interaction with the electrons in the solid will make, after some typical time τ , this electron to decay to a lower energy state of momentum \mathbf{p}' and energy E' , exciting one of the electrons in the solid from a state \mathbf{k} to \mathbf{k}' (See Fig. 1). From a perturbative approach one can write this lifetime (inverse of the probability per unit time) using Fermi's golden rule as done in Ref. 22 arriving at the expression

$$\frac{1}{\tau_\sigma} = \frac{2\pi}{\hbar} \int_{E_F}^E dE' \rho_\sigma^>(E') \int_0^{E-E'} d\varepsilon [\rho_\sigma^<(\varepsilon) \rho_\sigma^>(\varepsilon + \omega) + \rho_\sigma^<(\varepsilon) \rho_\sigma^>(\varepsilon + \omega)] |M(\omega)|^2, \quad (1)$$

where $\omega \equiv E - E'$ is the energy transfer and

$$\begin{aligned} \rho_\sigma^>(E') &= [1 - f(E')] \rho_\sigma(E') \\ \rho_\sigma^<(E') &= f(E') \rho_\sigma(E'), \end{aligned} \quad (2)$$

being $f(E)$ the Fermi-Dirac occupation function and $\rho_\sigma(E)$ the DOS for energy E and spin σ . $\bar{\sigma}$ is the opposite spin to σ . $M(\omega)$ is the matrix element of the screened electronic interaction that connects the initial and final states. To arrive at Eq. (1) we have made an angular average^{29,30} and the interference between direct and exchange terms has been neglected since they appear to be negligible when exchange effects are also taken into account in the interaction.³¹

In a nonmagnetic material $\rho_\sigma = \rho_{\bar{\sigma}}$ so the spin subindex can be dropped in Eq. (1) and

$$\frac{1}{\tau} = \frac{2\pi}{\hbar} \int_{E_F}^E dE' \rho^>(E') \int_0^{E-E'} d\varepsilon \rho^<(\varepsilon) \rho^>(\varepsilon + \omega) |M(\omega)|^2, \quad (3)$$

where now ρ is the spin-independent density of states. We will in our study make the approximation of constant matrix elements, because we are mainly interested in the direct effect of the DOS. One can suppose that, in the small energy range we are interested in (0–4 eV above E_F), matrix elements will remain fairly constant while in the same energy range the DOS could change about one order of magnitude. This constant matrix approximation has been widely used in the analysis of photoemission data with excellent results, see for example the pioneering work by Berglund and Spicer²⁹ where experimental photoemission results in copper and silver were successfully explained using constant matrix elements. Kane³⁰ calculated the matrix elements in the case of silicon obtaining that they were quite insensitive to energy changes (less than 30% when going from 5 to 8 eV). In the study of secondary electron cascades in ferromagnets, although energy-dependent matrix elements were necessary to obtain full numerical agreement with the experiment, the general trend was successfully explained using constant matrix elements.²² Also in ferromagnets, after removing from the scattering amplitudes for Stoner processes the contribution of the DOS, a relatively small energy dependence is found.³²

Once the matrix element has been extracted out of the integral, only the convolution integral of the DOS corresponding to the electrons involved in the deexcitation process has to be calculated. From Eq. (3) taking a constant value $\rho(E) = \rho_s$ for the DOS and assuming constant matrix elements, one arrives immediately at the familiar $(E - E_F)^{-2}$ scaling of the lifetime,

$$\frac{1}{\tau(E)} = \frac{\pi}{\hbar} \rho_s^3 |M|^2 (E - E_F)^2, \quad (4)$$

an expression that was first obtained in Ref. 29. By comparison with the well-known Quinn's result,¹¹ valid for free electrons in the low-energy limit ($E \approx E_F$), the matrix element is given by

$$|M|^2 = \frac{\sqrt{3}}{64} \frac{p_F}{p} \frac{\hbar \omega_p}{\rho_s^3 E_F^2} \left[\frac{2p_F q_{TF}}{(2p_F)^2 + q_{TF}^2} + \tan^{-1} \left(\frac{2p_F}{q_{TF}} \right) \right] \quad (5)$$

$$\approx \frac{\sqrt{3} \pi}{128} \frac{1}{\rho_s^3} \frac{\hbar \omega_p}{E_F^2} \propto n^{-11/6}. \quad (6)$$

The strength of the interaction decreases with increasing density due to the enhancement of the screening, thus increasing the lifetimes. It can be shown that in free-electron-like metals $\tau \propto n^{5/6}$ in the high-density limit.

In order to describe noble metals, a more detailed description of the density of states will be necessary. We will then distinguish between s and d electrons, describing them in different ways. Noble metals are characterized by a quite flat sp band and large d bands lying below the Fermi energy. The distance from E_F to the d band is about 2 eV in the case of Cu and near 4 eV in the case of Ag. It is not difficult to

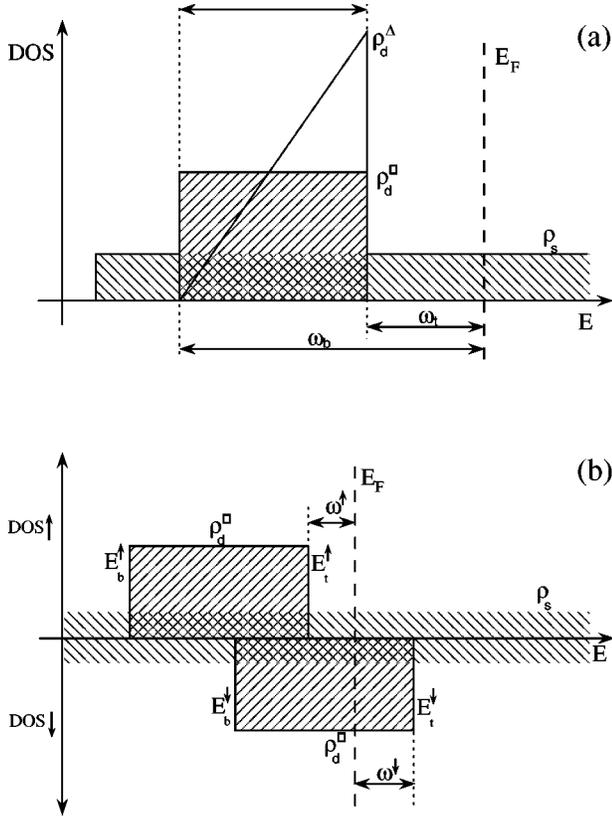


FIG. 2. (a) Box and triangle description for the DOS in noble metals. Observe that $\rho_d^{\text{triangle}} = 2\rho_d^{\text{box}}$ in order to keep the same number of electrons. (b) Box model for the spin dependent DOS in Co. We will assume that ρ_d and ρ_s are spin independent, therefore, the ferromagnetic behavior will be a consequence of the positions relative to the Fermi energy of the d boxes.

use first-principles calculations of band structure to perform the integral in Eq. (3) but, in order to understand the qualitative behavior, we have used simple descriptions for the DOS, which allow to obtain analytical results. In this way, we describe the DOS of noble metals using boxes with different shapes. We use a square box of height ρ_s for the sp electrons, extended over the whole energy range and another one for the d electrons, of height ρ_d , between E_b and E_t [see Fig. 2(a)]. Thus, we introduce

$$\rho(E) = \rho_s + \rho_d \theta(E_t - E) \theta(E - E_b). \quad (7)$$

In Eq. (7) $\theta(x)$ is the unit step function. Although the matrix elements are taken as constants we distinguish them by the participating electrons because one would expect that the overlap between different wave functions will be different, and hence yield different matrix elements.³³ $|M_{l_f s_f}^{l_i s_i}|^2$ is the matrix element of when the primary electron goes from band l_i to l_f and the secondary electron goes from band s_i to s_f . We suppose that $|M_{k_l}^{ij}|^2 = |M_{j_l}^{ki}|^2 = |M_{l_k}^{ii}|^2$ due to time-reversal symmetry and indistinguishable electrons, respectively.

When $(E - E_F) < \omega_t$, being $\omega_t \equiv (E_F - E_t)$ the energy distance from the top of the d band to the Fermi energy, no d electrons can be excited and the expression shown in Eq. (4) is reproduced,

$$\frac{1}{\tau(E)} = \frac{\pi}{\hbar} \rho_s^2 \rho_d \left(\frac{\rho_s}{\rho_d} \right) |M_{ss}^{ss}|^2 (E - E_F)^2. \quad (8)$$

When $\omega_t < (E - E_F) < \omega_b$ ($\omega_b \equiv E_F - E_b$), part of the d electrons can be excited and the lifetime is given by

$$\frac{1}{\tau(E)} = \frac{\pi \rho_s^2 \rho_d}{\hbar} \left[\left(\frac{\rho_s}{\rho_d} \right) |M_{ss}^{ss}|^2 (E - E_F)^2 + |M_{ss}^{sd}|^2 (E - E_F - \omega_t)^2 \right]. \quad (9)$$

Due to the new channel for the decaying provided by the d electrons, an additional contribution appears, which has not the standard $(E - E_F)^{-2}$ behavior predicted by the Fermi Liquid's theory in the case of low energies. This new term scales with $(E - E_F - \omega_t)^2$ and results in a sudden increase of the probability (decrease of the lifetime) as soon as it becomes possible to excite d electrons. Note also that the s -electron contribution scaling with $(E - E_F)^{-2}$ enters with a factor $[(\rho_s/\rho_d) \ll 1]$ thus being smaller than the d -electron contribution only considering DOS. As a result, and supposing as a first approximation that matrix elements are of the same order, once d electrons can take part in the de-excitation process, they dominate it while the s -electron contribution becomes a small correction. Finally for energies large enough for all d electrons to be excited, namely $(E - E_F) > \omega_b$, the term due to excitation of d electrons becomes a constant and only the s electron part keeps the energy dependence.

One can also use a triangular DOS for the d electrons [see Fig. 2(a)]

$$\rho_d(E) = \frac{E - E_b}{E_t - E_b} \rho_d \theta(E_t - E) \theta(E - E_b). \quad (10)$$

Following the same procedure, below the d -electron excitation threshold ($E - E_F < \omega_t$) the result in Eq. (8) remains unchanged. If part of the d electrons can be excited ($\omega_t < E - E_F < \omega_b$), defining $W \equiv E_t - E_b = \omega_b - \omega_t$,

$$\frac{1}{\tau(E)} = \frac{\pi \rho_s^2 \rho_d}{\hbar} \left\{ \left(\frac{\rho_s}{\rho_d} \right) |M_{ss}^{ss}|^2 (E - E_F)^2 + |M_{ss}^{sd}|^2 \left[(E - E_F - \omega_t)^2 - \frac{1}{3W} (E - E_F - \omega_t)^3 \right] \right\}. \quad (11)$$

Although the result is in this case more complicated, there is still a term related with the excitation of d electrons that has a energy dependence different from the $(E - E_F)^2$ behavior. Finally, when all d electrons can be excited ($\omega_b < E - E_F$) the d term becomes again a constant. Keep in mind that when using triangles, in order to keep the number of electrons constant, ρ_d in Eqs. (10) and (11) is twice ρ_d entering in Eqs. (7)–(9).

These analytic results have been compared with numeric calculations of the convolution integrals where first-principles density of states has been used. In order to compare both approaches, when evaluating the analytic expressions we will use for ρ_s the value given by first-principles

calculation at the Fermi energy.³⁴ To determine ρ_d we first calculate the total contribution of the d electrons by integration of the d part of the first-principles density of states and then we divide it by the d band width. (ρ_s/ρ_d) is approximately 0.07 in Cu and 0.09 in Ag, being small parameters in both cases. In spite of the huge simplification made in the DOS, the agreement found between our simplified model and the first-principles evaluation is very good, supporting the validity of our approach. A triangle gives the best agreement in the case of copper while a square box is more suitable for silver. These shapes are intuitively suggested by the appearance of the first-principles DOS.

III. LIFETIME IN FERROMAGNETIC METALS: COBALT

Ferromagnetic metals are characterized by different band structures for each spin state. This implies different DOS for each spin and so one expects different lifetimes too. Regarding the way of introducing this spin-dependent DOS, one will have to go back to Eq. (1) in which the spin label appears explicitly. The DOS in these materials are characterized by completely filled d bands for the majority spin state (\uparrow), which means that d^\uparrow bands are below the Fermi energy. Nevertheless, minority d bands are partially filled, crossing the Fermi energy. This implies that there exist d^\downarrow states above the Fermi energy. One expects that these empty d^\downarrow states will yield an increase of the available phase space for the decaying process leading to shorter lifetimes for spin-down electrons.²²

In the same way as we did for noble metals, we will describe the majority electrons by a constant value for the ρ_s^\uparrow and a box below Fermi energy for the ρ_d^\uparrow . In the case of minority spins the d box will cross the Fermi energy [See Fig. 2(b)]. Then we have

$$\rho^{\uparrow,\downarrow}(E) = \rho_s + \rho_d \theta(E_t^{\uparrow,\downarrow} - E) \theta(E - E_b^{\uparrow,\downarrow}), \quad (12)$$

where $E_b^\uparrow < E_t^\uparrow < E_F$ and $E_b^\downarrow < E_F < E_t^\downarrow$. For simplicity, it has been assumed that $\rho_s^\uparrow = \rho_s^\downarrow = \rho_s$ and $\rho_d^\uparrow = \rho_d^\downarrow = \rho_d$; this means that the whole bulk polarization comes from the differences in E_b^\uparrow and E_t^\uparrow . Introducing these expressions into Eq. (1), it is possible to obtain analytic results in the whole energy range but the expression becomes more and more involved while increasing the energy and so we will restrict ourselves to the lowest energy regions described below. In the same way as happened in the case of noble metals, the model using boxes is able to reproduce all the main features of a full first-principles calculation.³⁵

In order to confront later the experimental data,¹⁰ we are going to choose the d threshold values $(E_b^{\uparrow,\downarrow}, E_t^{\uparrow,\downarrow})$ in such a way that $\omega^\uparrow < \omega^\downarrow$, being $\omega^\uparrow \equiv (E_F - E_t^\uparrow)$ and $\omega^\downarrow \equiv (E_t^\downarrow - E_F)$, respectively the energy distances to the top of the majority and minority d bands, because this is the case for Co ($\omega^\uparrow \approx 0.6$ eV, $\omega^\downarrow \approx 1.3$ eV).³⁵ With this selection of parameters, the first energy range is $(E - E_F) < \omega^\uparrow$. In this region, the primary electron cannot excite any majority d electrons. Nevertheless, minority d electrons will be excited for any energy. Equation (1) for τ^\uparrow then gives

$$\frac{1}{\tau^\uparrow(E)} \approx \frac{2\pi\rho_d^3}{\hbar} \frac{1}{2} \left(\frac{\rho_s}{\rho_d} \right) |M_{sd}^{sd}|^2 (E - E_F)^2, \quad (13)$$

where the fact that a majority electron above the Fermi energy is always an s electron has been taken into account. Only the matrix elements entering with the lowest power in (ρ_s/ρ_d) have been considered because in Co $(\rho_s/\rho_d) \approx 0.2$. Notice that this approximation does not affect the energy dependence of the lifetime, only the prefactor of this dependence. In the case of minority electrons both spins are possible in this energy range, but we will consider that the electron above the Fermi level will be a d^\downarrow electron because ρ_d is much bigger than ρ_s . Thus, we obtain,

$$\frac{1}{\tau^\downarrow(E)} \approx \frac{2\pi\rho_d^3}{\hbar} \frac{1}{2} |M_{dd}^{dd}|^2 (E - E_F)^2. \quad (14)$$

In this energy range the ratio between lifetimes is constant and its value obviously depends both on the DOS and on the matrix elements. To lowest order in (ρ_s/ρ_d) we find

$$\frac{\tau^\uparrow}{\tau^\downarrow} \approx \frac{\rho_d}{\rho_s} \frac{|M_{dd}^{dd}|^2}{|M_{sd}^{sd}|^2}. \quad (15)$$

Notice that this ratio remains energy independent if higher order terms in (ρ_s/ρ_d) are included in Eqs. (13) and (14). For Co this result extends from the Fermi energy up to $\omega^\uparrow \approx 0.6$ eV.

The next energy region is characterized by $\omega^\uparrow < (E - E_F) < \omega^\downarrow$, where both minority and majority d electrons will be excited. In this case,

$$\frac{1}{\tau^\uparrow(E)} \approx \frac{2\pi\rho_d^3}{\hbar} \left[\frac{1}{2} \left(\frac{\rho_s}{\rho_d} \right) |M_{sd}^{sd}|^2 (E - E_F)^2 + \frac{1}{2} \left(\frac{\rho_s}{\rho_d} \right)^2 |M_{ss}^{sd}|^2 (E - E_F - \omega^\uparrow)^2 \right], \quad (16)$$

and

$$\frac{1}{\tau^\downarrow(E)} \approx \frac{2\pi\rho_d^3}{\hbar} \left[\frac{1}{2} |M_{dd}^{dd}|^2 (E - E_F)^2 + \left(\frac{\rho_s}{\rho_d} \right) |M_{ds}^{dd}|^2 (E - E_F - \omega^\uparrow)^2 \right]. \quad (17)$$

As can be seen, similar to the case of copper, there appear new terms that do not scale as $(E - E_F)^2$. These new terms change the constant ratio between τ^\uparrow and τ^\downarrow , which will now depend on energy. Due to the fact that the new term entering in τ^\downarrow has a smaller power in (ρ_s/ρ_d) than the one in τ^\uparrow , the ratio $(\tau^\uparrow/\tau^\downarrow)$ is expected to increase with increasing energy. Above this energy range $(E - E_F > 1.3$ eV) the expressions are too complicated to be of any use to spell out.

IV. ANALYSIS OF THE EXPERIMENTAL DATA

Information on excited-electron lifetime has been obtained from the measure of linewidths in photoemission and inverse photoemission experiments. A recent compilation of these experiments³⁶ suggests a linear dependence $\tau^{-1} = \alpha \cdot (E - E_F)$ with a universal value for α . Ballistic electron emission microscopy³⁷ (BEEM) seems also to be able to al-

low the determination of the mean-free path of very low-energy electrons. The lifetimes obtained for Au by BEEM (Ref. 38) can be well described by Quinn's approach while in the case of palladium an unexplained linear dependence with $(E - E_F)$ is obtained.³⁹ This linear dependence can be explained in materials where a large density of unoccupied states exists just above the Fermi energy, for instance in transition metals,²² but the quadratic behavior should be retrieved for energies low enough.

Nor linewidth analysis, where approximations for the elastic width are needed to separate the inelastic linewidth, neither BEEM, where the lifetime enters as one of the fitting parameters, provide a direct measure of the lifetime of the excited states. TR-2PPE does it. In a TR-2PPE experiment an electron is excited above the Fermi level but below the work function using an ultrashort laser pulse. A second pulse extracts the electron above the vacuum level and it can be detected. Measuring the final energy, the energy corresponding to the intermediate excited state can be determined simply by subtracting the photon energy. Varying the delay between the two laser pulses the time evolution of the population of this state, and hence its lifetime, can be *directly* monitored.

There are experimental results for the lifetime of low-energy electrons obtained by TR-2PPE in different groups.³⁻¹⁰ There exist problems related to the interpretation of the data and there is presently a lack of agreement between the results of different groups. Different processes could be the origin of this disagreement. First of all, the deexcitation of the primary electrons (and their corresponding holes) excited by the laser beam gives rise to a series of secondary cascade processes that would affect the measured lifetime by a delayed repopulation of the states under measure. This kind of process would mainly affect the states up to about 1 eV above the Fermi energy. When the energy of the laser is enough to excite electrons from the deeper d bands the corresponding d holes are localized and presumably have lifetimes longer than those of the free states that could be the reason for some surprising results.^{8,9} Transport effects would also affect lifetimes by taking the excited electrons out from the experimental region. This transport effects would depend on the quality of the sample and on its orientation and could explain the observed discrepancies and the face-dependent lifetimes observed by some groups.^{6,7} Most probably, the discrepancies reflect different experimental approaches (like different pulse-width or different strategies of data analysis) and/or a considerably different quality of the used samples. See Ref. 40 for a recent discussion on this question. Thus, even if TR-2PPE experiments do not measure the intrinsic lifetime of the excited state but a superposition of the different processes involved in the photoexcitation event, we think that an analysis of the experimental data in terms of our approach could be useful to check the consistency of both experimental and theoretical results.

Copper is the material with most available data; we have used four different sources.⁵⁻⁸ We have fitted the experimental data to the expression obtained for a triangular density of states of the d band, which gives the best agreement with the first-principles calculation,

TABLE I. Fitting parameters for the different experimental sources. a_0 accounts for the term with usual $(E - E_F)^2$ scaling and a_1 for the one scaling with $(E - E_F - \omega_t)^2$ [see Eqs. (18) and (21)]. Units are [$\text{fs}^{-1} \text{eV}^{-2}$].

Data	a_0		a_1
	Ag	Cu	Cu
Ref. 5	4.2×10^{-2}	0.8×10^{-2}	0.10
Ref. 6	5.7×10^{-2}	2.4×10^{-2}	
Ref. 7		0.8×10^{-2}	1.6×10^{-2}
Ref. 8		2.2×10^{-2}	
Quinn (Ref. 11)	5.5×10^{-2}	4.1×10^{-2}	
Quinn (ϵ_0) (Ref. 13)	3.0×10^{-2}	1.7×10^{-2}	

$$\frac{1}{\tau} = a_0(E - E_F)^2 + a_1 \left[(E - E_F - \omega_t)^2 - \frac{1}{3W}(E - E_F - \omega_t)^3 \right] \theta(E - E_F - \omega_t), \quad (18)$$

where $\omega_t = 1.9 \text{ eV}$ and $W \approx 4.1 \text{ eV}$ from band-structure calculations.³⁴ a_0 and a_1 are directly related to the matrix elements [c.f. Eqs. (8) and (11)],

$$a_0 = \frac{\pi}{\hbar} \rho_s^2 \rho_d \left(\frac{\rho_s}{\rho_d} \right) |M_{ss}^{ss}|^2 \quad a_1 = \frac{\pi}{\hbar} \rho_s^2 \rho_d |M_{ss}^{sd}|^2. \quad (19)$$

The experimental lifetimes are reasonably well described by Eq. (18), which supports the validity of the constant matrix approximation that has been used. The results from our fittings are summarized in Table I. As can be seen the values for a_0 look rather consistent but there is a large spread in the values for a_1 . However, it is in the region where a_1 plays a role where the biggest experimental errors are expected because lifetimes are much shorter and closer to the experimental resolution. Regarding the ratio between the matrix elements for Cu, it varies between

$$1.1 < \frac{|M_{ss}^{ss}|}{|M_{ss}^{sd}|} < 2.8. \quad (20)$$

One would expect, supposing the screening is the same for all electrons, that this ratio should in fact be greater than unity due to a bigger overlap between the s -wave functions.³³

For silver, we have two sources^{4,5} and due to the large d band threshold there is no experimental information of the behavior above that threshold. We cannot, therefore, obtain information about a_1 . The available experimental data has been fitted using the expression

$$\frac{1}{\tau} = a_0(E - E_F)^2. \quad (21)$$

The a_0 values obtained are shown in Table I. Since Cu and Ag have very similar ρ_s values, the difference in the a_0 values has to be related to the matrix elements and the screening. To first order, we expect the bare matrix elements to be rather similar but the screening is expected to be less efficient in Ag

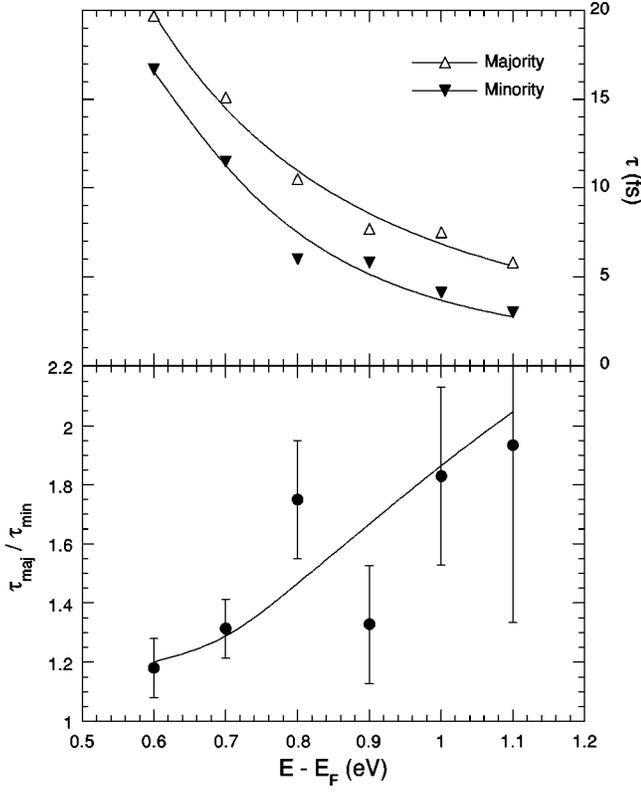


FIG. 3. Spin-dependent lifetimes in Cobalt (top). Open triangles are experimental data for majority spin electrons while full triangles correspond to minority spin electrons (Ref. 10). The solid lines are the results of our model applied to both sets of data. The ratio between the spin-dependent lifetimes (bottom) is also compared with our results.

than in Cu (in Ag, d bands are deeper and ρ_s is smaller) and hence $a_0(\text{Ag})$ should be larger than $a_0(\text{Cu})$ as found (see Table I).

Aeschlimann *et al.*¹⁰ have measured spin-dependent lifetimes in Co in the energy range we have studied. We have analyzed both spin states with the expression

$$\frac{1}{\tau_{\uparrow\downarrow}} = a_0^{\uparrow\downarrow} (E - E_F)^2 + a_1^{\uparrow\downarrow} (E - E_F - \omega_{\uparrow})^2 \theta(E - E_F - \omega_{\uparrow}), \quad (22)$$

from the previous section. Figure 3 shows the fitting of the available experimental data for both spin states together with the ratio of the lifetimes. The model seems to be able to provide a good description of the experiment, which supports the validity of the approximations being used. To lowest order in (ρ_s/ρ_d) ,

$$a_0^{\uparrow} \approx \frac{\pi}{\hbar} \rho_d^3 \left(\frac{\rho_s}{\rho_d} \right) |M_{sd}^{sd}|^2 \quad a_0^{\downarrow} \approx \frac{\pi}{\hbar} \rho_d^3 |M_{dd}^{dd}|^2. \quad (23)$$

This means that for energies where no d^{\uparrow} electrons can be excited the lifetimes will be dominated by the excitation of d^{\downarrow} electrons giving a contribution scaling with $(E - E_F)^2$.

TABLE II. Fitting parameters for Co. a_0 accounts for the term with usual $(E - E_F)^2$ scaling and a_1 for the one scaling with $(E - E_F - \omega_{\uparrow})^2$ for both majority (\uparrow) and minority (\downarrow) spin states [see Eq. (22)]. Units are [$\text{fs}^{-1} \text{eV}^{-2}$].

	$a_0[\text{Co}]$		$a_1[\text{Co}]$	
Data	\uparrow	\downarrow	\uparrow	\downarrow
Ref. 10	0.14	0.17	0.04	0.65

When excitation of d^{\uparrow} electrons becomes possible there appears a correction, which scales with $(E - E_F - \omega_{\uparrow})^2$. Again to lowest order in (ρ_s/ρ_d) ,

$$a_1^{\uparrow} \approx \frac{\pi}{\hbar} \rho_d^3 \left(\frac{\rho_s}{\rho_d} \right)^2 |M_{ss}^{sd}|^2 \quad a_1^{\downarrow} \approx \frac{\pi}{\hbar} \rho_d^3 \left(\frac{\rho_s}{\rho_d} \right) |M_{ds}^{dd}|^2. \quad (24)$$

Notice that, in contrast to the case of noble metals, a_1 has a higher power in (ρ_s/ρ_d) than a_0 and hence, the correction is supposed to be small.

The best-fitting parameters are shown in Table II. It is interesting to notice that $a_0^{\uparrow\downarrow}(\text{Co})$ is about one order of magnitude larger than $a_0(\text{Cu})$ and $a_0(\text{Ag})$. This is a consequence of the existence of empty minority d states above the Fermi energy, which increases the available phase space with respect to the case of noble metals. The absence of these states in the case of majority electrons is also the key to understand why a_1^{\uparrow} is about one order of magnitude smaller than the other parameters. The same argument would predict a larger value for a_0^{\downarrow} . This is probably not found due to a small overlap between the localized d states.³³

Finally, from the different ratios between the $a_{1,2}^{\uparrow\downarrow}$ the ratios between the different matrix elements can be obtained, showing that

$$|M_{dd}^{dd}| < |M_{sd}^{sd}| < |M_{ss}^{sd}| < |M_{ds}^{dd}|. \quad (25)$$

Except for the case $|M_{ds}^{dd}|$ the matrix elements follow the intuitive trend that the more d electrons the smaller the matrix elements should be, which is supported by previous results.³³

V. SUMMARY

We have developed a model that introduces the effect of the real density of states in the calculation of the lifetime of low-energy electrons in transition metals, in a simple way. With the assumption of constant matrix elements and using them as fitting parameters, different sets of experimental data can be accounted for. The values obtained for the matrix elements presently differ between different experiments on the order of 40% and show a qualitative agreement with theoretical estimations. Mechanisms affecting the measurements such as transport, cascades or surface effects can be the reason for this disagreement. On the other hand, energy-dependent matrix elements and interference terms should also be taken into account, but this is beyond the present initial analysis.

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