

First-principles multiple scattering theory of quasiparticle lifetimes in ferromagnetic materials

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Abstract

We develop a first-principles approach for evaluating the lifetimes of excited electrons in Fe and Ni based on the self-energy formalism of many-body theory. The GW approximation is applied for the lowest self-energy term whereas the high-order terms are evaluated within T -matrix approach. Both for Ni and Fe we find important contributions of the T -matrix self-energy terms corresponding to the excited electron scattering from spin waves.

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The electron scattering processes in ferromagnetic materials have been studied by means of photoemission spectroscopy [1] and spin-dependent time-resolved two-photon photoemission spectroscopy (TR-2PPE) [2]. The lifetimes, linewidths and mean free paths of excited electrons were a subject of theoretical researches [2,3]. An important aspect of the dynamics of hot electron damping in these materials is the role of spin-flip processes including Stoner excitations and spin-wave emission. In our study, we tackle this problem by a first-principles method based on the self-energy formalism of many-body theory.

We calculate the excited electron lifetimes from the imaginary part of the self-energy operator expectation value [4]. The polarization function, dielectric function and screened potential are evaluated within RPA approach. The first-order term of the self-energy is calculated within GW approximation [4]. We incorporate the spin-flip processes by invoking an ab initio T -matrix formalism [5] where the T -matrix operator is

defined as a solution of Bethe–Salpiter equation

$$\begin{aligned} \mathcal{T}_{\sigma_1, \sigma_2}(1, 2|3, 4) = & \mathcal{W}(1, 2)\delta(1-3)\delta(2-4) \\ & + \mathcal{W}(1, 2) \int d1' d2' \mathcal{K}_{\sigma_1, \sigma_2}(1, 2|1', 2') \\ & \times \mathcal{T}_{\sigma_1, \sigma_2}(1', 2'|3, 4). \end{aligned} \quad (1)$$

For the screened potential W we apply static approximation. The two-particle propagator K is a product of two one-particle spin-dependent Green functions

$$\mathcal{K}_{\sigma_1, \sigma_2}(1, 2|1', 2') = \mathcal{G}_{\sigma_1}(1, 1')\mathcal{G}_{\sigma_2}(2', 2). \quad (2)$$

The direct term of the T -matrix contribution to the self-energy is expressed as

$$\Sigma_{\sigma_2}^d(4, 2) = - \sum_{\sigma_1} \int d1 d3 \mathcal{G}_{\sigma_1}(3, 1)\mathcal{T}_{\sigma_1, \sigma_2}(1, 2|3, 4). \quad (3)$$

With $\sigma_1 = \sigma_2$ we have high-order contributions to the non-spin-flip scattering, with $\sigma_1 \neq \sigma_2$ this term provides spin-flip Stoner and spin-wave contribution to self-energy. The pole frequencies of T -matrix correspond to magnon energies. We have derived the explicit equations for self-energy Σ^d in frequency representation and performed numerical calculations based on LMTO band-structure method [6].

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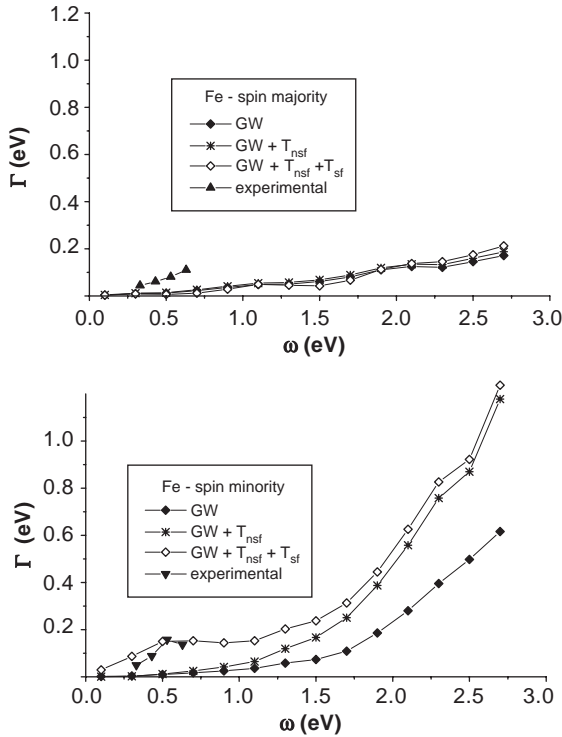


Fig. 1. Momentum-averaged linewidths of excited electrons in Fe. GW means the GW contributions to the linewidths whereas T_{nsf} and T_{sf} are, respectively, non-spin-flip and spin-flip contributions of the T -matrix theory.

In Figs. 1 and 2 we show the momentum-averaged linewidths Γ of electron excitations in Fe and Ni in frequency range accessible for the TR-2PPE measurements. For both we find that the inclusion of T -matrix terms markedly affects the Γ values and lifetimes $\tau = (\hbar/2\pi)\Gamma^{-1}$. In particular, big are differences between Γ for spin-minority excited electrons in Fe calculated within GW approximation and within GW + T method. At low frequencies the $\Gamma(\text{GW} + T)$ line widths are 5–7 times more than $\Gamma(\text{GW})$ values. Thus we conclude that the spin-wave generation, which is allowed for the damping of spin-minority electrons by the total spin-moment conservation law, effectively decreases the lifetimes. For de-excitation of spin-minority electrons in Fe the inclusion of T -matrix terms greatly improves the agreement between calculated and experimental data [2]. For the spin-minority electrons of Fe we also obtain a noticeable increase of Γ at frequencies above 1.5 eV related to the high-order non-spin-flip effects. For spin-

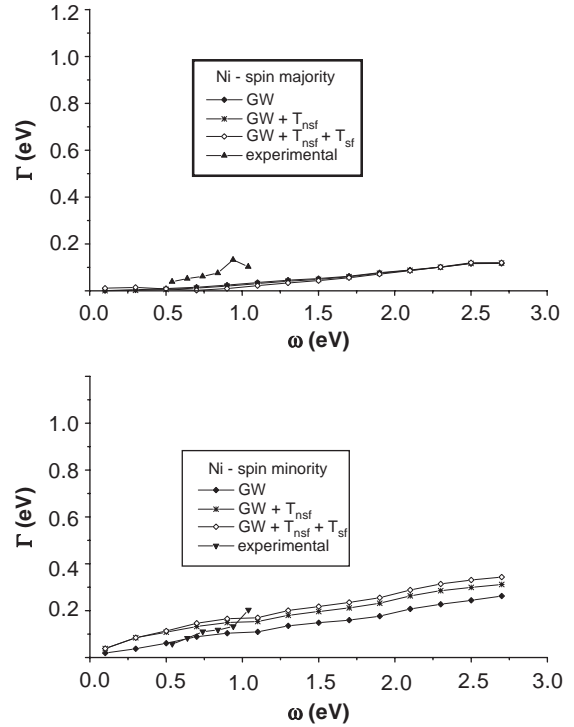


Fig. 2. Momentum-averaged linewidths of excited electrons in Ni. Notations are the same as in Fig. 1.

majority excited electrons in Fe and Ni we notice, however, big differences between experimental and theoretical data. We refer these differences to the effects not included in our theoretical model, in particular, cascade electrons effects, transport and Auger excitations effects which were discussed in Ref. [2].

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