

# Non-quasiparticle states in the core level spectra of ferromagnetic semiconductors and half-metallic ferromagnets

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**Abstract.** The Green's functions that determine X-ray spectra are calculated in the  $s-d$  exchange model of a saturated conducting ferromagnet in the presence of the core hole. It is demonstrated that the core level (X-ray absorption, emission and photoelectron) spectroscopy might be an efficient tool to investigate the nonquasiparticle (NQP, spin-polaron) states in electron energy spectrum since the core hole potential can enhance essentially their spectral weight. NQP contributions to resonant X-ray scattering spectra can be also much more pronounced than those to the the density of states.

**PACS.** 78.70.Dm X-ray absorption spectra – 78.70.En X-ray emission spectra and fluorescence – 75.50.Pp Magnetic semiconductors – 75.50.Cc Other ferromagnetic metals and alloys

## 1 Introduction

Connections between magnetic and electronic properties of materials are very important both conceptually and from the point of view of possible applications. Half-metallic ferromagnets (HMF) [1–3], ferromagnetic semiconductors [4–6], and colossal magnetoresistance materials [7–9] are of special interest for spintronics (spin-dependent electronics) [10]. Core level spectroscopy techniques such as X-ray absorption, X-ray emission, and photoelectron spectroscopies (xas, xes, and xps, correspondingly) give an essential information about the electronic structure of these substances (see, e.g., references [11–14]). Unfortunately, a physical background to understand corresponding experimental data is still very poor. First, theoretical description of core-level spectra is a complicated problem because of core hole effects [15, 16]. Second, analysis of the spectra is often based on the naive one-particle picture of density of states (DOS), in particular, on the old Stoner picture of magnetism formation. The latter theory is fully inapplicable for real itinerant magnets where effects of electron correlations play a crucial role. These effects are especially important in HMF which present a limiting case of ultimately strong itinerant magnets [2].

Presence of the energy gap for the band states with one spin projection results in a considerable contribution of the non-quasiparticle (NQP) states which can occur in the gap [2, 17–20]. The formation of NQP states is a consequence of formation of a spin polaron, i.e., a superposition of minority-spin conduction electron states with the states of majority-spin conduction electron *plus* magnon excitations. This effect has purely quantum nature (it disappears in the limit of large localized spins) and essentially many-body character. In particular, the singularities in the electron density of states owing to NQP contributions are pinned to the Fermi level, similar to the standard Kondo effect. Recently, a first-principle calculation of the NQP states was performed for NiMnSb [21]. The incoherent NQP states can exhibit themselves in the tunneling phenomena, see references [22–24]. Moreover, it is demonstrated in these papers that they determine  $I-V$  characteristics of tunnel junctions in the absence of normal quasiparticle transport, which is the case in the presence of the energy gap. Therefore they are directly related to spintronics. Besides that, the NQP states can be observed in the photoelectron spectroscopy of conduction band [18, 20, 25] and in the nuclear magnetic relaxation [2, 26].

Effects of the NQP states in the core-level spectroscopy have been recently discussed qualitatively for CrO<sub>2</sub> in reference [13]. Being almost currentless, these states are in a sense strongly localized in the real space. Therefore

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they yield a contribution to the elastic peak of X-ray fluorescence, which is typical for insulators, but usually absent for metals. In this work we present a quantitative microscopic theory which gives a basis for the calculation of NQP contributions to various core level spectra: xas, xes, xps and X-ray resonant scattering.

The corresponding two-particle Green's functions which determine the spectral properties via Kubo formulas are treated in Section 2. It turns out to be that in this case three-body problem (conduction electron, magnon, and core hole) can be solved in a compact closed form suitable for calculations with any bare density of electron states. To demonstrate qualitative effects, we present in Sections 3–5 the results of simple numerical calculations of various core level spectra for the model semielliptic DOS. We show that usually NQP contributions to core-level spectra are strongly enhanced in comparison with those to initial DOS and therefore X-ray spectroscopy might be a suitable tool to investigate them.

## 2 Calculation of two-particle Green's functions

To consider the core level problem we use the Hamiltonian of  $s-d$  exchange model in the presence of external potential  $U$  induced by the core hole:

$$\mathcal{H} = \sum_{\mathbf{k}\sigma} t_{\mathbf{k}} c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}\sigma} + \varepsilon_0 f^{\dagger} f - U \sum_{\mathbf{k}\mathbf{k}'\sigma} c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}'\sigma} f^{\dagger} f - I \sum_{\mathbf{q}\mathbf{k}} \sum_{\alpha\beta} \mathbf{S}_{\mathbf{q}} c_{\mathbf{k}\alpha}^{\dagger} \sigma_{\alpha\beta} c_{\mathbf{k}-\mathbf{q}\beta} + \sum_{\mathbf{q}} J_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} \mathbf{S}_{-\mathbf{q}}. \quad (1)$$

Here  $c_{\mathbf{k}\sigma}^{\dagger}$ ,  $c_{\mathbf{k}\sigma}$  and  $\mathbf{S}_{\mathbf{q}}$  are operators for conduction electrons and localized spins in the quasimomentum representation,  $f^{\dagger}$ ,  $f$  are core hole creation and annihilation operators,  $t_{\mathbf{k}}$  is the band energy,  $I$  is the parameter of the  $s-d$  exchange interaction which is assumed to be local,  $\sigma$  are the Pauli matrices,  $J_{\mathbf{q}}$  are the Fourier transforms of the exchange parameters between localized spins, which determine spin dynamics. The  $s-d$  exchange model, where the systems of local moments and current carriers are separated, describes a ferromagnetic metal or degenerate semiconductor. Provided that the band filling ( $E_F$  calculated from the bottom) is smaller than the spin splitting  $\Delta = 2|I|S$ , this is the simplest model of HMF [2].

We use the method of double-time retarded Green's functions [27]. The Green's function for operators  $A$  and  $B$

$$\langle\langle A|B \rangle\rangle_E^{\pm} = \int_{-\infty}^0 dt e^{iEt} \langle [e^{i\mathcal{H}t} A e^{-i\mathcal{H}t}, B]_{\pm} \rangle, \quad \text{Im } E > 0 \quad (2)$$

satisfies the equation of motion

$$E \langle\langle A|B \rangle\rangle_E^{\pm} = \langle [A, B]_{\pm} \rangle + \langle\langle [A, \mathcal{H}] | B \rangle\rangle_E^{\pm} \quad (3)$$

$$E \langle\langle A|B \rangle\rangle_E^{\pm} = \langle [A, B]_{\pm} \rangle + \langle\langle A | [\mathcal{H}, B] \rangle\rangle_E^{\pm}. \quad (4)$$

We write down the equation of motion (3) for the commutator two-particle Green's function

$$G_{\mathbf{k}\mathbf{k}'}^{\sigma}(E) = \langle\langle c_{\mathbf{k}\sigma} f | f^{\dagger} c_{\mathbf{k}'\sigma}^{\dagger} \rangle\rangle_E \quad (5)$$

which takes into account core-level hole effects and determines X-ray absorption and emission spectra [15]. For the ferromagnetic state at low temperatures (in the spin-wave region) we can pass to the magnon representation for the spin operators,

$$S_i^z = S - b_i^{\dagger} b_i, \\ S_i^{\pm} = (2S)^{1/2} b_i, \quad S_i^{-} = (2S)^{1/2} b_i^{\dagger}.$$

Then we derive (the electron energy  $E$  is referred to  $\varepsilon_0$ )

$$(E - t_{\mathbf{k}\sigma}) G_{\mathbf{k}\mathbf{k}'}^{\sigma}(E) = (1 - n_f - n_{\mathbf{k}}^{\sigma}) \left[ \delta_{\mathbf{k}\mathbf{k}'} - U \sum_{\mathbf{p}} G_{\mathbf{p}\mathbf{k}'}^{\sigma}(E) \right] - I \Phi_{\mathbf{k},\mathbf{k}'}^{\sigma}(E) \quad (6)$$

where  $t_{\mathbf{k}\sigma} = t_{\mathbf{k}} - \sigma I \langle S^z \rangle$  is the Hartree-Fock spectrum,  $n_{\mathbf{k}}^{\sigma} = n(t_{\mathbf{k}\sigma})$  is the Fermi function,  $n_f$  is the occupation number for the  $f$ -hole in the initial state, which is further on will be put to zero. We will take into account the occupation numbers  $n_{\mathbf{k}}^{\sigma}$  in a simple ladder approximation which works well at small enough concentrations of current carriers, except for the immediate vicinity of the Fermi edge. One should note that the ladder approximation for the  $s-d$  exchange model is not the same as for the Hubbard model, but is much better owing to a proper treatment of localized electron spins. For the Hubbard model without core hole our approximation is equivalent to the Edwards-Hertz approach [17] which provides an adequate description of saturated ferromagnetic state in a broad range of conduction electron concentrations. At the same time, we do not treat here the problem of the X-ray edge singularity where more advanced approaches are necessary [15,16].

We use in equation (6) the notation

$$\Phi_{\mathbf{k}-\mathbf{p},\mathbf{k}'}^{\sigma}(E) = \sum_{\mathbf{r}} F_{\mathbf{k}-\mathbf{p}-\mathbf{r},\mathbf{r},\mathbf{k}'}^{\sigma}(E) \quad (7)$$

$$F_{\mathbf{k}-\mathbf{p},\mathbf{q},\mathbf{k}'}^{\sigma}(E) = (2S)^{1/2} \langle\langle b_{\mathbf{q}}^{\sigma} c_{\mathbf{k}-\mathbf{p},-\sigma} f | f^{\dagger} c_{\mathbf{k}'\sigma}^{\dagger} \rangle\rangle_E, \quad (8) \\ b_{\mathbf{q}}^{\pm} = b_{-\mathbf{q}}^{\dagger}, \quad b_{\mathbf{q}}^{-} = b_{\mathbf{q}}.$$

The Green's function  $F$  satisfies the equation

$$(E - t_{\mathbf{k}-\mathbf{p},-\sigma} + \sigma\omega_{\mathbf{q}}) F_{\mathbf{k}-\mathbf{p},\mathbf{q},\mathbf{k}'}^{\sigma}(E) = -U(1 - n_{\mathbf{k}-\mathbf{p}}^{-\sigma}) \Psi_{\mathbf{q},\mathbf{k}'}^{\sigma}(E) - I(N_{\mathbf{q}}^{\sigma} + \sigma n_{\mathbf{k}-\mathbf{p}}^{-\sigma}) \times [2SG_{\mathbf{k}-\mathbf{p}+\mathbf{q},\mathbf{k}'}^{\sigma}(E) + \sigma\Phi_{\mathbf{k}-\mathbf{p}+\mathbf{q},\mathbf{k}'}^{\sigma}(E)] \quad (9)$$

where we have performed decouplings in spirit of the ladder approximation,

$$\sum_{\mathbf{k}''} \langle\langle c_{\mathbf{k}''-\sigma}^{\dagger} c_{\mathbf{k}''+\mathbf{q}\sigma} c_{\mathbf{k}-\mathbf{p},-\sigma} f | f^{\dagger} c_{\mathbf{k}'\sigma}^{\dagger} \rangle\rangle_E \rightarrow -n_{\mathbf{k}-\mathbf{p}}^{-\sigma} G_{\mathbf{k}-\mathbf{p}+\mathbf{q},\mathbf{k}'}^{\sigma}(E),$$

$\omega_{\mathbf{q}}$  is the magnon frequency,  $\langle b_{\mathbf{q}}^{\sigma} b_{\mathbf{q}}^{-\sigma} \rangle = N_{\mathbf{q}}^{\sigma} = \sigma N(\sigma\omega_{\mathbf{q}})$ ,  $N(\omega) = 1/[\exp(\omega/T) - 1]$  is the Bose function,

$$\Psi_{\mathbf{q},\mathbf{k}'}^{\sigma}(E) = \sum_{\mathbf{r}} F_{\mathbf{k}-\mathbf{r},\mathbf{q},\mathbf{k}'}^{\sigma}(E). \quad (10)$$

For  $U = 0$  we have

$$G_{\mathbf{k}\mathbf{k}'}^{\sigma}(E) = (1 - n_{\mathbf{k}}^{\sigma})\delta_{\mathbf{k}\mathbf{k}'}G_{\mathbf{k}}^{\sigma}(E) \quad (11)$$

where  $G_{\mathbf{k}}^{\sigma}(E)$  is the one-electron Green's function of the ideal crystal,

$$G_{\mathbf{k}}^{\sigma}(E) = [E - t_{\mathbf{k}\sigma} - \Sigma_{\mathbf{k}}^{\sigma}(E)]^{-1}, \quad \Sigma_{\mathbf{k}}^{\sigma}(E) = \frac{2I^2 \langle S^z \rangle Q_{\mathbf{k}}^{\sigma}}{1 + \sigma I Q_{\mathbf{k}}^{\sigma}} \quad (12)$$

with

$$Q_{\mathbf{k}}^{\uparrow}(E) = \sum_{\mathbf{q}} \frac{N_{\mathbf{q}} + n_{\mathbf{k}+\mathbf{q}}^{\downarrow}}{E - t_{\mathbf{k}+\mathbf{q}\downarrow} + \omega_{\mathbf{q}}},$$

$$Q_{\mathbf{k}}^{\downarrow}(E) = \sum_{\mathbf{q}} \frac{1 + N_{\mathbf{q}} - n_{\mathbf{k}-\mathbf{q}}^{\uparrow}}{E - t_{\mathbf{k}-\mathbf{q}\uparrow} - \omega_{\mathbf{q}}}.$$

Note that equation (12) yields correctly the exact Green's function in the limit of an empty conduction band at  $T = 0$  [20,28,29]. The result (12) corresponds to the approximation [17] in the theory of strong itinerant ferromagnetism (which is just the case of half-metallic ferromagnets, cf. [19]). This approximation can be justified with the use of a Ward identity. Such an approximation in the  $s-d$  exchange model is also widely used for metals [30].

For  $I = 0$  one obtains

$$G_{\mathbf{k}\mathbf{k}'}^{\sigma}(E) = \frac{1 - n_{\mathbf{k}}^{\sigma}}{E - t_{\mathbf{k}\sigma}} \left[ \delta_{\mathbf{k}\mathbf{k}'} - \frac{UP^{\sigma}(E)}{1 + UP^{\sigma}(E)} \right] \quad (13)$$

$$P^{\sigma}(E) = \sum_{\mathbf{k}} \frac{1 - n_{\mathbf{k}}^{\sigma}}{E - t_{\mathbf{k}\sigma}}. \quad (14)$$

In the general case, we have a three-particle problem (conduction electron, core hole and magnon) which requires a careful mathematical investigation. However, we can use the fact that the magnon frequencies are much smaller than typical electron energies and, last but not least, that the resolution of xas and xes methods is not sufficient to probe the energy scale of a typical magnon frequency so we can put the latter to zero. Neglecting spin dynamics the equations (6), (9) can be solved exactly in a rather simple way provided that we consider the case of zero temperatures ( $N_{\mathbf{q}}^{+} = 0, N_{\mathbf{q}}^{-} = 1$ ). Under these conditions  $Q$  does not depend on quasimomenta, and the scattering by the core hole in the presence of a magnon is described by the same  $\mathbf{q}$ -independent resolvent  $P$ . We derive from equation (9)

$$\Phi_{\mathbf{k}-\mathbf{p}+\mathbf{q},\mathbf{k}'}^{\sigma}(E) = -\frac{1}{1 + \sigma I Q^{\sigma}(E)} \left[ 2ISQ^{\sigma}(E)G_{\mathbf{k}-\mathbf{p}+\mathbf{q},\mathbf{k}'}^{\sigma}(E) + UP^{-\sigma}(E)\Psi_{\mathbf{q},\mathbf{k}'}^{\sigma}(E) \right] \quad (15)$$

On summing over  $\mathbf{p}$  we see that  $\Psi_{\mathbf{q},\mathbf{k}'}^{\sigma}$  does not depend on  $\mathbf{q}$ ,

$$\Psi_{\mathbf{q},\mathbf{k}'}^{\sigma}(E) = \Psi_{\mathbf{k}'}^{\sigma}(E) = (2S)^{1/2} \langle \langle b_{\mathbf{c}-\sigma}^{\dagger} f | f^{\dagger} c_{\mathbf{k}'\sigma}^{\dagger} \rangle \rangle_E$$

$$= \sum_{\mathbf{p}\mathbf{q}} F_{\mathbf{k}-\mathbf{p},\mathbf{q},\mathbf{k}'}^{\sigma}(E) = \sum_{\mathbf{p}} \Phi_{\mathbf{k}-\mathbf{p},\mathbf{k}'}^{\sigma}(E) \quad (16)$$

(physically it means that the electron and magnon operator should belong to the same perturbed site). Then equation (15) can be solved in terms of  $\Psi$  to obtain

$$\Psi_{\mathbf{k}'}^{\sigma}(E) = -\frac{2ISQ^{\sigma}(E)}{1 + UP^{-\sigma}(E) + \sigma I Q^{\sigma}(E)} R_{\mathbf{k}'}^{\sigma}(E), \quad (17)$$

$$R_{\mathbf{k}'}^{\sigma}(E) = \sum_{\mathbf{k}} G_{\mathbf{k}\mathbf{k}'}^{\sigma}(E). \quad (18)$$

After substituting equation (17) into equation (15), summing over  $\mathbf{p} = \mathbf{q}$  and substituting the result into equation (6) we obtain the closed equation for the Green's function  $G$

$$[E - t_{\mathbf{k}\sigma} - \Sigma^{\sigma}(E)] G_{\mathbf{k}\mathbf{k}'}^{\sigma}(E) = (1 - n_{\mathbf{k}}^{\sigma}) \left[ \delta_{\mathbf{k}\mathbf{k}'} - U \sum_{\mathbf{p}} G_{\mathbf{p}\mathbf{k}'}^{\sigma}(E) \right] - \frac{U \Sigma^{\sigma}(E) P^{-\sigma}(E)}{1 + UP^{-\sigma}(E) + \sigma I Q^{\sigma}(E)} \sum_{\mathbf{p}} G_{\mathbf{p}\mathbf{k}'}^{\sigma}(E).$$

Neglecting the factors  $(1 - n_{\mathbf{k}}^{\sigma})$  which is possible at small band filling (anyway, they are irrelevant for our purposes), we have the standard result for the impurity scattering (13) with the renormalized energy spectrum  $E_{\mathbf{k}\sigma} = t_{\mathbf{k}\sigma} + \Sigma^{\sigma}(E)$  and the effective impurity potential

$$U_{ef}^{\sigma}(E) = U \left[ 1 + \frac{\Sigma^{\sigma}(E) P^{-\sigma}(E)}{1 + UP^{-\sigma}(E) + \sigma I Q^{\sigma}(E)} \right]. \quad (19)$$

Then the local density of states is given by

$$N_{\text{loc}}^{\sigma}(E) = -\frac{1}{\pi} \text{Im} G_{00}^{\sigma}(E) \quad (20)$$

with

$$G_{00}^{\sigma}(E) = \sum_{\mathbf{k}\mathbf{k}'} G_{\mathbf{k}\mathbf{k}'}^{\sigma}(E) = R_{\sigma}(E) + T^{\sigma}(E) R_{\sigma}^2(E)$$

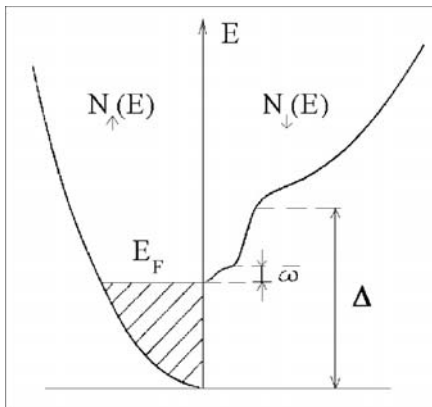
$$= \frac{R_{\sigma}(E)}{1 + U_{ef}^{\sigma}(E) R_{\sigma}(E)} \quad (21)$$

where

$$R_{\sigma}(E) = G_{00}^{(0)\sigma}(E) = \sum_{\mathbf{k}} G_{\mathbf{k}}^{\sigma}(E), \quad (22)$$

$G_{\mathbf{k}}^{\sigma}(E)$  is given by equation (12), and the  $T$ -matrix has the form

$$T^{\sigma}(E) = -\frac{U_{ef}^{\sigma}(E)}{1 + U_{ef}^{\sigma}(E) R_{\sigma}(E)}. \quad (23)$$



**Fig. 1.** Density of states in a half-metallic ferromagnet with  $I > 0$  (schematically). Non-quasiparticle states with  $\sigma = \downarrow$  occur above the Fermi level.

### 3 X-ray absorption and emission spectra

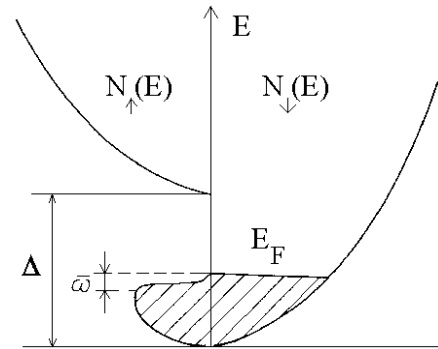
Generally speaking, theoretical investigation of core level spectra requires a numerical calculations with account of realistic bandstructure. We restrict ourselves to some model examples. The resolvents in the complex plane can be calculated analytically for the simple semielliptical bare density of states (see Appendix).

The picture of the NQP contributions to the DOS of an ideal crystal is described by the Green's function (12). This was considered in references [18,20] for a degenerate ferromagnetic semiconductor and discussed in detail for a half-metallic ferromagnet [2]. Remember that the energy gap occurs in the case where  $\Delta = 2|I|S > E_F$ . We have

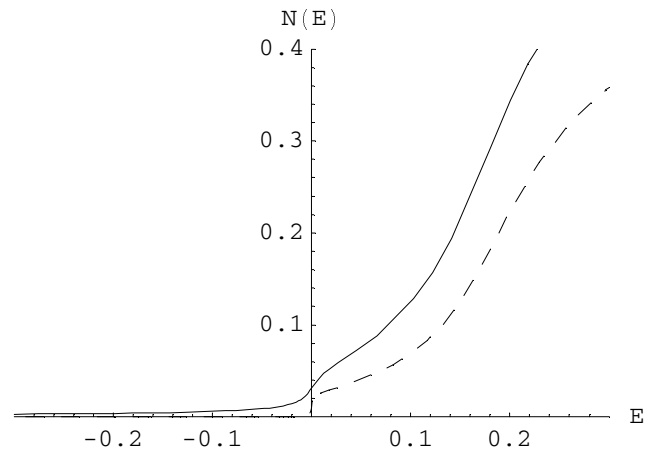
$$\delta N^\sigma(E) = -\frac{1}{\pi} \text{Im} R_\sigma(E) = -\frac{1}{\pi} \text{Im} \Sigma(E) |R'_\sigma(E)|. \quad (24)$$

As demonstrates an analysis of the electron-magnon interaction, the picture turns out to be different for two possible signs of the  $s-d$  exchange parameter  $I$ . For  $I < 0$  spin-up NQP states are present below the Fermi level as an isolated region (Fig. 1): occupied states with the total spin  $S - 1/2$  are a superposition of the states  $|S\rangle|\downarrow\rangle$  and  $|S-1\rangle|\uparrow\rangle$ . On the contrary, for  $I > 0$  the spin-down NQP scattering states form a “tail” of the upper spin-down band, which starts from  $E_F$  (Fig. 2) since the Pauli principle prevents the electron scattering into occupied states. Of course, the jumps at the Fermi level are in fact smeared at the scale of the magnon energies  $\bar{\omega}$ .

Since xas probes empty states and xes occupied states, the quantity (20) describes the absorption spectrum for  $E > E_F$  and emission spectrum for  $E < E_F$ . As follows from above results, the picture observed in the core level spectroscopy is determined by more complicated integral equations in comparison with the DOS problem. Therefore some new circumstances occur for the NQP contributions. The numerical results in our simple model are shown in Figures 3–4, the local density of states for  $U = 0$  (which



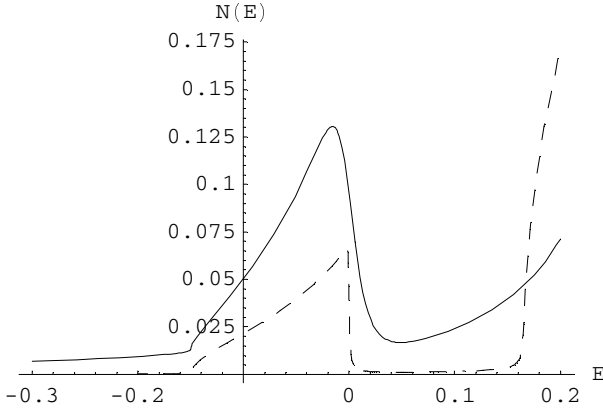
**Fig. 2.** Density of states in a half-metallic ferromagnet with  $I < 0$  (schematically). Non-quasiparticle states with  $\sigma = \uparrow$  occur below the Fermi level.



**Fig. 3.** The local density of states  $N_{loc}^\downarrow(E)$  (solid line) for a half-metallic ferromagnet with  $S = 1/2$ ,  $I = 0.3$ ,  $\delta = 0.01$  in the presence of the core hole potential  $U = 0.2$ . The dashed line shows the DOS  $N_1(E)$  for the ideal crystal with spin dynamics being neglected. The value of  $E_F$  calculated from the band bottom is 0.15. The energy  $E$  is referred to the Fermi level.

coincides with usual DOS in that case) being also presented for comparison. To take into account core level broadening, we introduce in the quantity (20) a finite damping  $\delta$  near the Fermi level (see Appendix).

For  $I > 0$  the results (19)–(21) provide full solution of the Kondo problem for an impurity in the ferromagnet, corresponding to the parquet approximation [31]. On the other hand, for  $I < 0$  the situation is complicated by the presence of the “false” Kondo divergence in the quantity (23), similar to the same approximation in the standard Kondo problem [32]. Formally, the  $T$ -matrix has a pole, and DOS above the Fermi level even turns out to be negative. Generally speaking, more advanced approximations are needed to solve this problem. However, this difficulty is not important for the X-ray problem where a large damping is always present, and experiments are performed at sufficiently high temperatures with rather poor resolution (as compared with a scale of the “Kondo temperature”). The case of very low temperatures and small



**Fig. 4.** The local density of states  $N_{\text{loc}}^{\uparrow}(E)$  (solid line) for a half-metallic ferromagnet with  $S = 1/2$ ,  $I = -0.3$ ,  $\delta = 0.025$  in the presence of the core hole potential  $U = 0.2$ . The dashed line shows the DOS  $N_{\uparrow}(E)$  for the ideal crystal. The value of  $E_F$  calculated from the band bottom is 0.15.

damping requires a special treatment which will be given elsewhere.

To leading order in  $U$  and  $I$  we obtain

$$\delta N_{\text{loc}}^{\sigma}(E) = \frac{1 - \text{Re}(U_{ef}^{\sigma}(E)/\Sigma^{\sigma}(E)) |R_{\sigma}^2(E)/R'_{\sigma}(E)|}{|1 + U_{ef}^{\sigma}(E)R_{\sigma}(E)|^2} \delta N^{\sigma}(E) - \frac{\text{Re}(U_{ef}^{\sigma}(E)/P^{-\sigma}(E)) |R_{\sigma}(E)|^2}{|1 + U_{ef}^{\sigma}(E)R_{\sigma}(E)|^2} \frac{1}{\pi} \text{Im} P^{-\sigma}(E). \quad (25)$$

The term in equation (25) with  $\text{Im} P^{-\sigma}(E)$  results in a smooth contribution to the spectrum. In particular, it is non-zero in the energy gap. Note that for the emission spectra such a term is absent. The NQP contributions to the absorption and emission spectra, that are proportional to  $\delta N^{\sigma}(E)$ , occur for  $I > 0$  and  $I < 0$  only, respectively.

The NQP contribution is the only minority contribution to  $\delta N^{\sigma}(E)$  near the Fermi level. As for majority contribution, this is smooth at the Fermi level and can be roughly treated in the Hartree-Fock approximation and obtained by a shift of the quasiparticle minority contribution by the spin splitting  $\Delta$  (Figs. 1, 2).

One can see from Figure 3 that the upturn of the NQP tail which occurs for  $I > 0$  becomes somewhat more sharp, although the jump near  $E_F$  weakens. For  $I < 0$  the spectral weight of NQP contributions increases in the presence of the core hole too (Fig. 4). These effects have a simple physical interpretation.

Since  $U_{ef}^{\sigma}(E) > 0$  and for small band filling  $R_{\sigma}(E) < 0$  near  $E_F$ , the denominator of the expression (25) results in a considerable enhancement of NQP contributions to the spectra in comparison with those to DOS. However, effects of interaction  $U$  do not reduce to a constant factor in the self-energy, but turn out to be non-trivial. Strong interaction with the core hole results in a deformation of conduction band. With increasing  $U$  the spectral density passes

to the band bottom. This effect is especially important for the NQP states since they lie in this region. Therefore the spectral weight of the NQP states increases. However, with further increasing  $U$  (at very large, in fact unrealistic values) a bound state is formed near the band bottom, and the NQP spectral weight becomes suppressed owing to factor of  $U$  in the denominator of the expression (19).

The temperature dependence of the spectra at relatively high temperatures can be roughly taken into account by neglecting short-range order, i.e., formally, by neglecting  $\mathbf{q}$ -dependence of spin correlation function. We have just to replace  $N_{\mathbf{q}} \rightarrow S - \bar{S}$  in the resolvent  $Q$ , so that effects of finite temperatures result in a decrease of the jumps and in an increase of NQP density of states in the energy gap.

## 4 Photoelectron spectroscopy

To discuss xps we consider the anticommutator core level Green's function

$$G_f(E) = \langle\langle f|f^{\dagger} \rangle\rangle_E = [E - \varepsilon_0 - \Sigma_f(E)]^{-1}. \quad (26)$$

By using both the equations (3) and (4) the self-energy  $\Sigma_f(E)$  is expressed in terms of an irreducible Green's function,

$$\Sigma_f(E) = U^2 \sum_{\sigma\sigma'} \langle\langle c_{\sigma}^{\dagger} c_{\sigma} f | f^{\dagger} c_{\sigma'}^{\dagger} c_{\sigma'} \rangle\rangle_E^{\text{irr}} \quad (27)$$

(the superscript “irr” means that the contributions divergent as  $(E - \varepsilon_0)^{-n}$  should be omitted in the equations of motion for the Green's function (27), cf. Ref. [20]).

To evaluate the Green's function (27) we pass to the time representation and use the simplest decoupling (the vertices are neglected),

$$\Sigma_f(E) = -U^2 \sum_{\sigma} \int_0^{\infty} dt e^{iEt} \langle c_{\sigma}^{\dagger}(t) c_{\sigma} \rangle \langle c_{\sigma}(t) f(t) f^{\dagger} c_{\sigma}^{\dagger} \rangle. \quad (28)$$

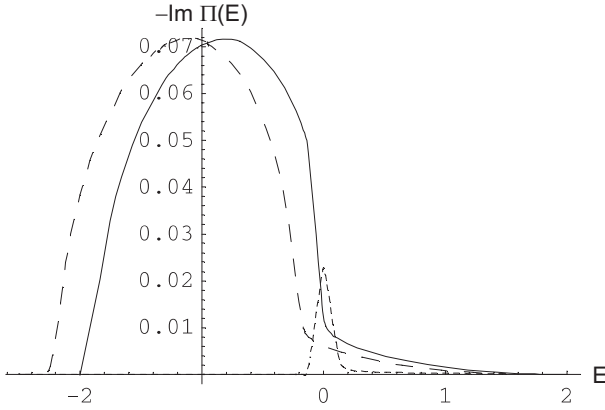
Using the spectral representation for the correlation functions of  $c$ -operators we obtain

$$\Sigma_f(E) = \frac{U^2}{\pi} \sum_{\sigma} \int dE' f(E') \text{Im} G_{00}^{(0)\sigma}(E') G_{00}^{\sigma}(E' - E). \quad (29)$$

Thus the photoelectron spectrum contains NQP contributions both from initial and final states, the hole effects being important only for final states. Photocurrent can be in principle resolved in spin projection of photoelectrons, which makes possible to observe the NQP contributions.

Since only the states below  $E_F$  are observed in xps, we have to treat spin up NQP states for  $I < 0$ . The leading quasiparticle contribution from spin down states reads

$$\delta \Sigma_{f\downarrow}(E) \simeq U^2 \sum_{\mathbf{k}\mathbf{k}'} \frac{n_{\mathbf{k}\downarrow}(1 - n_{\mathbf{k}'\downarrow})}{E - t_{\mathbf{k}\downarrow} + t_{\mathbf{k}'\downarrow}} = U^2 \Pi_0(E). \quad (30)$$



**Fig. 5.** The functions  $-\text{Im } \Pi_0(E)$  (solid line),  $-\text{Im } \Pi_1(E)$  (dashed line) and  $-\text{Im } \Pi_2(E)$  (short-dashed line) for a half-metallic ferromagnet with  $S = 1/2$ ,  $I = -0.3$ ,  $\delta = 0.01$ . The value of  $E_F$  calculated from the band bottom is 0.15.

Despite of absence of spin up band states (at least, in our simple model of HMF), transitions in NQP states with  $\sigma = \uparrow$  (see Fig. 2) take place owing to electron-magnon scattering. The corresponding NQP contribution has the structure

$$\delta\Sigma_{f\uparrow}(E) = \delta\Sigma_{f\uparrow}^{(1)}(E) + \delta\Sigma_{f\uparrow}^{(2)}(E) \quad (31)$$

with

$$\delta\Sigma_{f\uparrow}^{(1)}(E) \propto -2I^2 S U^2 R_{\uparrow}'(E) \Pi_1(E), \quad (32)$$

$$\Pi_1(E) = \sum_{\mathbf{k}\mathbf{k}'} \frac{n_{\mathbf{k}\downarrow}}{E - t_{\mathbf{k}\downarrow} + t_{\mathbf{k}'\uparrow}} \quad (33)$$

$$\delta\Sigma_{f\uparrow}^{(2)}(E) \propto (2I^2 S)^2 U^2 [R_{\uparrow}'(E)]^2 \Pi_2(E), \quad (34)$$

$$\Pi_2(E) = \sum_{\mathbf{k}\mathbf{k}'} \frac{n_{\mathbf{k}\downarrow} n_{\mathbf{k}'\downarrow}}{E - t_{\mathbf{k}\downarrow} + t_{\mathbf{k}'\downarrow}}. \quad (35)$$

To leading order in  $I$  only initial NQP states make a contribution. The term  $\delta\Sigma_{f\uparrow}^{(1)}(E)$  has a threshold corresponding to the energy gap, but is smeared practically over the whole band (Fig. 5). The high-order contribution  $\delta\Sigma_{f\uparrow}^{(2)}(E)$  starts from small  $E$  and is more singular,

$$\text{Re } \delta\Sigma_{f\uparrow}^{(2)}(E) \propto E \ln |E|, \quad (36)$$

$$\text{Im } \delta\Sigma_{f\uparrow}^{(2)}(E) \propto E|E|. \quad (37)$$

## 5 Resonant X-ray scattering

Now we consider NQP effects in resonant X-ray scattering processes. It was observed recently [13] that the elastic peak of the X-ray scattering in  $\text{CrO}_2$  is observed which is more pronounced than in usual Cr compounds, e.g., the elemental chromium. The authors of this work have put forward some qualitative arguments that the NQP states may give larger contributions to resonant X-ray scattering than usual itinerant electron states. Here we shall treat

this question quantitatively and estimate explicitly the corresponding enhancement factor. The intensity of resonant X-ray emission induced by the photon with the energy  $\omega$  and polarization  $q$  is given by the Kramers-Heisenberg formula [12, 33, 34]

$$\mathcal{I}_{q'q}(\omega', \omega) \propto \sum_n \left| \sum_l \frac{\langle n | C_{q'} | l \rangle \langle l | C_q | 0 \rangle}{E_0 - \omega' - E_l - i\Gamma_l} \right|^2 \times \delta(E_n + \omega' - E_0 - \omega). \quad (38)$$

Here  $q', \omega'$  are the polarization and energy of the emitted photon,  $|n\rangle, |0\rangle$  and  $|l\rangle$  are the final, initial and intermediate states of the scattering system, respectively,  $E_i$  are the corresponding energies,  $C_q$  is the operator of the dipole moment for the transition, which is proportional to  $fc + c^\dagger f^\dagger$ . For simplicity we will assume hereafter that  $\Gamma_l$  does not depend on the intermediate state,  $\Gamma_l = \Gamma$ , and take into account only the main X-ray scattering channel where the hole is filled from the conduction band (for a more general multichannel consideration, see, e.g., Ref. [35]). Assuming also that the electron-photon interaction that induces the transition is contact, the expression for the threshold scattering intensity can be obtained from (38) in the form [35]

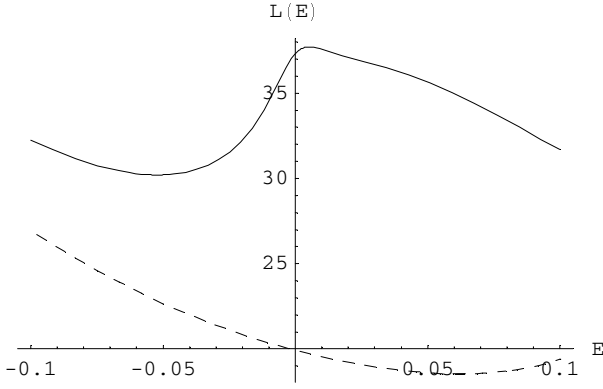
$$\mathcal{I}_{\omega'} \propto \sum_{\sigma\sigma'} \int_0^\infty dt_1 \int_0^\infty dt_2 \times \exp[-i(\omega' - E_0)(t_1 - t_2) - \Gamma(t_1 + t_2)] \times \langle 0 | c_\sigma \exp(i\mathcal{H}_f t_1) c_{\sigma'}^\dagger, \exp[i\mathcal{H}_i(t_2 - t_1)] c_{\sigma'} \times \exp(-i\mathcal{H}_f t_2) c_\sigma^\dagger | 0 \rangle \quad (39)$$

where  $H_f$  and  $H_i$  are conduction-electron Hamiltonians with and without core hole, respectively. The complicated correlation function in (39) can be decoupled in the ladder approximation which is exact for the empty conduction band. Then we obtain [35]

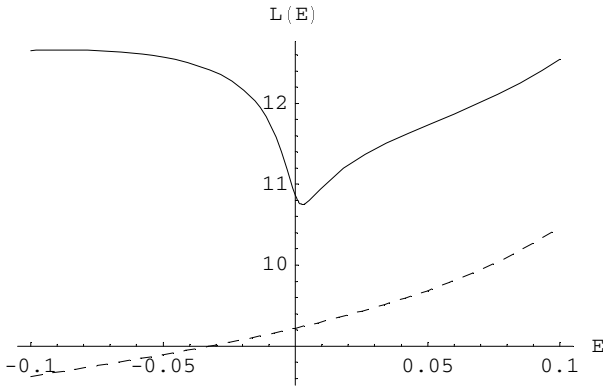
$$\mathcal{I}_{\omega'} \propto W^2 L(\omega' - E_0), \quad (40)$$

$$L(E) = \left| \sum_\sigma G_{00}^\sigma(E + i\Gamma) \right|^2$$

where  $G_{00}^\sigma$  is given by (21),  $W$  is a transition matrix element (in a simple model of Ref. [35] this is imaginary part of the hole potential). Owing to a jump in the density of states at the Fermi level, the NQP part of the Green's function contains a large logarithm  $\ln(D/[\omega' - E_0 + i\Gamma])$  at small  $|\omega' - E_0|$ ,  $D$  being a bandwidth. It means that the corresponding contribution to the elastic X-ray scattering intensity ( $\omega' = E_0$ ) is enhanced by a factor of  $\ln(D/\Gamma)$ . Provided that the NQP contribution dominates over the majority quasiparticle contribution (which is possible for small  $\Gamma$  only), the enhancement factor in  $\mathcal{I}_{\omega'}$  is  $\ln^2(D/\Gamma)$ . This makes a quantitative estimation for the qualitative effect discussed in reference [13]. Of course, smearing of the jump in the NQP density of states by spin dynamics



**Fig. 6.** The function  $L(E)$  with account of NQP contributions (solid line), and in the Hartree-Fock approximation (dashed line) for a half-metallic ferromagnet with  $S = 1/2$ ,  $I = 0.3$ ,  $\Gamma = 0.01$ . The value of  $E_F$  calculated from the band bottom is 0.15, the hole potential is  $U = 0.3$ .



**Fig. 7.** The function  $L(E)$  with account of NQP contributions (solid line), and in the Hartree-Fock approximation (dashed line) for a half-metallic ferromagnet with  $S = 1/2$ ,  $I = -0.3$ ,  $\Gamma = 0.01$ . The value of  $E_F$  calculated from the band bottom is 0.15, the hole potential is  $U = 0.1$ .

is irrelevant provided that  $\Gamma \gtrsim \bar{\omega}$  ( $\bar{\omega}$  is a characteristic magnon frequency).

Figures 6–7 show the function  $L(E)$  for both signs of the  $s-d$  exchange parameter  $I$ . One can see that NQP contributions result in a maximum at  $E = 0$  for  $I > 0$  and in a minimum for  $I < 0$ . The difference is due to different signs of the NQP jumps at the Fermi level and, consequently, of the corresponding logarithms in the real part. The numerical results are also presented for the Hartree-Fock approximation (where the Green's functions  $G_{\mathbf{k}}^{\sigma}(E) = (E - t_{\mathbf{k}\sigma})^{-1}$  are substituted into (21), (40)) which does not take into account NQP effects. The deviation turns out to be asymmetric because the NQP contributions are asymmetric with respect to the Fermi level. Of course, our calculation is not quite strict (in particular, the NQP terms in  $W$  should be in principle considered). However, this corresponds to account of main singular contributions.

## 6 Conclusions and discussion

To conclude, we emphasize the main points of the present work. First, the non-quasiparticle (NQP) states (in HMF these are the only minority states which are present near the Fermi level) do manifest themselves in the core level spectroscopy. This demonstrates an important role of correlation effects beyond mean field (or Fermi-liquid) picture in metallic magnets (for a general discussion, see Ref. [2]). Moreover, due to interference of interactions of electrons with magnons and with core holes an enhancement of NQP contributions is possible in comparison with the initial density of states.

From a formal point of view, the results obtained in Section 2 yield a non-trivial analytical solution of a three-body problem. For the case  $I > 0$  which takes place for the most of known “spintronic” materials (including colossal magnetoresistance manganites) the “ladder” approximation provides probably a complete solution describing basic physics for both semiconductors and metals. For the case  $I < 0$  some analogue of the Kondo effect occurs which may lead to complicated theoretical issues. It is not too important pragmatically since in the spectroscopy problems the Kondo-like divergences are cut at the inverse core level lifetime  $\Gamma$ . However, further theoretical investigations are of interest to obtain a more consistent scheme.

To describe HFM state, we used in our calculations a simple model of saturated ferromagnet (Figs. 1–2). However, the numerical results can change substantially for more realistic bandstructure of HMF. A smooth enough band structure is expected, e.g., for such strong itinerant ferromagnets as pyrite systems  $\text{Fe}_{1-x}\text{Co}_x\text{S}_2$ , see references [37] (according to these electronic structure calculations,  $\text{CoS}_2$  is an almost HMF system). On the other hand, hybridization peaks near the energy gap exist in the HMF from the class of the Heusler alloys and in  $\text{CrO}_2$  (see Ref. [2]). Thus calculations with the use of a concrete electron structure of HMF are needed for a detailed comparison with X-ray spectra, which can be done on the basis of equations obtained in the present paper. A more simple situation is expected in some ferromagnetic semiconductors, e.g., chalcogenide spinels  $\text{HgCr}_2\text{Se}_4$  and  $\text{CdCr}_2\text{Se}_4$  [38]. The conduction band in  $\text{HgCr}_2\text{Se}_4$  has a spherical symmetry. In the paramagnetic phase the valence band is fourfold degenerate at the point  $\Gamma$ , and below the Curie point the equal-energy surfaces are transformed into ellipsoids due to exchange field. In  $\text{CdCr}_2\text{Se}_4$  the bottom of  $s$ -like conduction band is at the point  $\Gamma$  (symmetry  $\Gamma_1$ ). Thus a parabolic spectrum model can be used directly for  $n$ -type spinels.

To probe the “spin-polaron” nature of the NQP states more explicitly, it would be desirable to use spin-resolved spectroscopical methods such as X-ray magnetic circular dichroism (XMCD, for a review see Ref. [36]). Owing to interference of electron-magnon scattering and “exciton” effects (interaction of electrons with the core hole) the NQP contributions to X-ray spectra can be considerably enhanced in comparison with those to DOS of the ideal crystal.

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## Appendix

We consider the resolvent

$$R(E) = \sum_{\mathbf{k}} \frac{n_{\mathbf{k}}}{E - t_{\mathbf{k}}} = \int_{-1}^{\mu} dE' \frac{\rho(E')}{E - E'} \quad (41)$$

for the semielliptic band with the width of  $D = 2$  and the bare density of states

$$\rho(E) = \frac{2}{\pi} \sqrt{1 - E^2}, \quad (42)$$

$\mu$  being the chemical potential (the Fermi energy). We have to calculate the function

$$R(z) = \frac{2}{\pi} \int_{-1}^{\mu} \frac{dE \sqrt{1 - E^2}}{z - E} = \frac{2}{\pi} \int_{-\mu}^1 \frac{dE \sqrt{1 - E^2}}{z + E}. \quad (43)$$

We make the substitution  $E = \cos \phi$ ,  $\phi \in (0, \pi/2)$ . Then  $\mu = -\cos \phi_0$  ( $\mu < 0$ ) and

$$\begin{aligned} R(z) &= \frac{2}{\pi} \int_0^{\phi_0} \frac{d\phi [1 - (z + \cos \phi - z)^2]}{z + \cos \phi} \\ &= \frac{2}{\pi} \left[ (1 - z^2) M(z) + z\phi_0 - \sqrt{1 - \mu^2} \right] \end{aligned} \quad (44)$$

where

$$M(z) = \int_0^{\phi_0} \frac{d\phi}{z + \cos \phi}. \quad (45)$$

On substituting  $\zeta = e^{i\phi}$ ,  $\zeta_0 = e^{i\phi_0}$  we obtain  $\mu = -\frac{1}{2}(\zeta_0 + 1/\zeta_0)$  or

$$\zeta_0 = -\mu + i\sqrt{1 - \mu^2}. \quad (46)$$

Then we have

$$\begin{aligned} M(z) &= -2i \int_1^{\zeta_0} \frac{d\zeta}{\zeta^2 + 2z\zeta + 1} \\ &= \frac{2i}{\zeta_1 - \zeta_2} \int_1^{\zeta_0} d\zeta \left( \frac{1}{\zeta - \zeta_2} - \frac{1}{\zeta - \zeta_1} \right) \end{aligned}$$

where

$$\zeta_{1,2} = -z \pm \sqrt{z^2 - 1}. \quad (47)$$

This gives us a desired complex function

$$M(z) = \frac{i}{\sqrt{z^2 - 1}} \left( \ln \frac{\zeta_0 - \zeta_2}{1 - \zeta_2} - \ln \frac{\zeta_0 - \zeta_1}{1 - \zeta_1} \right). \quad (48)$$

Note that the standard choice of the branch of logarithmic function,  $\ln z = \ln |z| + i \arg(z)$ , corresponds to the advanced Green's function. To consider the retarded Green's function, we have to change the sign of the imaginary part. On the real axis we obtain

$$\text{Im } R(E) = \begin{cases} -2\sqrt{1 - E^2}, & -1 < E < \mu \\ 0, & \text{otherwise} \end{cases} \quad (49)$$

$$\begin{aligned} \text{Re } R(E) &= \frac{2}{\pi} \left[ (1 - E^2) M_1(E) \right. \\ &\quad \left. - \sqrt{1 - \mu^2} + E(\pi/2 + \arcsin \mu) \right]. \end{aligned} \quad (50)$$

For  $|E| > 1$  we have

$$M_1(E) = \frac{2}{E} (1 - 1/E^2)^{-1/2} \arctan \left[ \frac{1 + u}{1 - u} \left( \frac{E - 1}{E + 1} \right)^{1/2} \right] \quad (51)$$

where

$$u = \mu / (1 + \sqrt{1 - \mu^2}).$$

For  $|E| < 1$  we derive

$$\begin{aligned} M_1(E) &= \frac{1}{\sqrt{1 - E^2}} \left[ \ln \left| \frac{Eu - 1 - \sqrt{1 - E^2}}{Eu - 1 + \sqrt{1 - E^2}} \right| \right. \\ &\quad \left. + \ln \left| \frac{E + 1 - \sqrt{1 - E^2}}{E + 1 + \sqrt{1 - E^2}} \right| \right]. \end{aligned} \quad (52)$$

The logarithmic terms can be smeared with a corresponding smearing of a jump in the real part,

$$\ln |x - a| \rightarrow \frac{1}{2} \ln [(x - a)^2 + \delta^2], \quad (53)$$

$$\theta(x) = \frac{1}{2} + \text{sign } x \rightarrow \frac{1}{2} + \frac{1}{\pi} \arctan \frac{x}{\delta}. \quad (54)$$

The integral

$$\tilde{R}(E) = \sum_{\mathbf{k}} \frac{1 - n_{\mathbf{k}}}{E - t_{\mathbf{k}}}$$

can be obtained as

$$\tilde{R}(E) = R(E)|_{\mu=1} - R(E).$$

On the real axis we have

$$\text{Re } R(E)|_{\mu=1} = \begin{cases} 2E, & |E| < 1 \\ 2[E - (E^2 - 1)^{1/2}], & |E| > 1. \end{cases} \quad (55)$$

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