The electronic damping of Eu and U chalcogenides

1989 J. Phys.: Condens. Matter 1 2631

(http://iopscience.iop.org/0953-8984/1/15/009)

J. Phys.: Condens. Matter **1** (1989) 2631–2636. Printed in the UK

The electronic damping of Eu and U chalcogenides

J M Wesselinowa
Department of Physics, University of Sofia, 5 Boulevard Anton Ivanov, 1126 Sofia, Bulgaria

Received 8 July 1988, in final form 13 December 1988

Abstract. The s–f model is used for investigating ferromagnetic f systems with ferromagnetic ($I > 0$) and antiferromagnetic ($I < 0$) s–f exchange interaction between conduction electrons and localised magnetic f moments. The dependence of the electronic damping on the ratio $|I/W|$ (where I is the s–f coupling constant and W the band width) for different temperatures is discussed. It is strikingly different in the strong-, intermediate- and weak-coupling regions. The role of the Coulomb interaction is shown.

1. Introduction

The so-called s–f (or d–f) model is widely accepted as a good description of magnetic 4f or 5f compounds (Nolting 1979, Reim 1986). The exchange coupling (s–f or d–f coupling) between the conduction band electrons and the lattice of localised magnetic moments which is the fundamental element in the theory of this model influences optical, electrical and magnetic properties of these materials. As first pointed out by Shastry and Mattis (1981), the physical properties depend very strongly on the sign of the s–f (or d–f) exchange. In the Eu chalcogenides as well as in Gd the electron spin couples ferromagnetically to the f spin (Nolting 1979), whereas in the actinide compounds such as the U chalcogenides the d–f coupling seems to be antiferromagnetic (AFM) (Reim 1986). The magnetic f–d exchange energy is established as negative for all the investigated metallic U compounds, i.e. the f and d moments align antiparallel on magnetic ordering. This property manifests itself in two magneto-optical effects. Firstly, the conduction electron spin polarisation displays a negative sign and it has a value of about -100% in UAs, about -40% in US and about -35% in USe but only about -20% in UTe. These results are in fair agreement with the results of spin-polarised photo-emission experiments (Erbudak and Meier 1980), which have shown the same sign and approximate size of σ_d and in particular the reduced σ_d for UTe. This negative conduction electron spin polarisation with different maximum values was also observed theoretically in our previous paper (Wesselinowa 1987). Secondly, the f $\rightarrow$ d transition energy displays a magnetic red shift, the size of which is found to depend on the sublattice magnetisation rather than on the net moment. Recently, Auslender and Bebenin (1988) have reported that the s–d and p–d exchange constants in the ferromagnetic (FM) semiconductors $CdCr_2Se_4$ and $HgCr_2Se_4$ are negative: $I = -0.05$ eV.

Nolting *et al* (1985) and Nolting and Dubil (1985) have discussed the electronic spectrum at $T = 0$ for different values of the ratio I/W (where I is the intra-atomic s–f

0953-8984/89/152631 + 06 $02.50

(or d–f) coupling and W is the band width) for FM and AFM systems, respectively. These researchers treat the electron damping phenomenologically through a Gaussian *ansatz* for the one-electron spectral density. They have not considered the Coulomb interaction, but our results show that it plays an important role and must be taken into account. In fact the imaginary parts of the energies corresponding to lifetime-broadening effects in the FM phase have not been considered until now. Recently, Karmakar *et al* (1986) have studied the electron damping rate $\gamma(\boldsymbol{k}=0)$ of the s–f model for $I>0$ in the paramagnetic phase close to T_C. In our previous papers (Wesselinowa 1984, 1987, hereafter referred to as I and II, respectively), using a Green function method proposed by Tserkovnikov (1971), we have calculated for the first time beyond the RPA the electronic spectrum and the damping of the s–f (or s–d) model with FM and AFM s–f exchange interaction between the conduction electrons and the localised magnetic moments. The properties are discussed for different parameters. The aim of the present paper is to obtain the dependence of the electronic damping on the ratio I/W for different temperatures ($T \neq 0$).

2. The model

For the description of the 4f and 5f systems we use the s–f (or d–f) model. The starting point is the following Hamiltonian:

$$H = H_M + H_E + H_{ME} \tag{1}$$

where H_M is the Heisenberg Hamiltonian for the ferromagnetically ordered f electrons:

$$H_M = -\tfrac{1}{2}\sum_{i,j} J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j - g\mu_B H \sum_i S_i^z. \tag{2}$$

$\boldsymbol{S}_i$ is the operator of the spin localised at the lattice site $\boldsymbol{R}_i$. The exchange interaction J_{ij} is a function of the lattice constant. H is an applied field in the z direction.

H_E represents the usual Hamiltonian of the conduction band electrons:

$$H_E = \sum_{q,\sigma} (\varepsilon_{q\sigma} - \mu) a_{q\sigma}^+ a_{q\sigma} + \tfrac{1}{2}\sum_{\substack{q,k',k'\\ \sigma,\sigma'}} v(\boldsymbol{q}) a_{k'-q\sigma}^+ a_{k'+q\sigma'}^+ a_{k'\sigma'} a_{k''\sigma} \tag{3}$$

where $a_{q\sigma}^+$ and $a_{q\sigma}$ are the Fermi creation and annihilation operators in the state $\boldsymbol{q}\sigma$, μ is the chemical potential, $\varepsilon_{q\sigma}$ are the Bloch energies given by

$$\varepsilon_{q\sigma} = \varepsilon_q - \sigma\mu_B H \qquad \sigma = \pm 1$$

and $v(\boldsymbol{q})$ is the Coulomb interaction.

The most important term in (1) is the operator H_{ME} which couples the two subsystems (2) and (3) by an intra-atomic exchange interaction:

$$H_{ME} = -\frac{I}{2N}\sum_{q,p} [S_{q-p}^+ a_{p-}^+ a_{q+} + S_{q-p}^- a_{p+}^+ a_{q-} + S_{q-p}^z (a_{p+}^+ a_{q+} - a_{p-}^+ a_{q-})] \tag{4}$$

where I is the constant interaction energy.

3. Static properties

To study the electronic excitation spectrum of the system, we shall evaluate the retarded Green function

$$G_{k\sigma}(t) = -i\theta(t)\langle[a_{k\sigma}(t); a_{k\sigma}^{+}]\rangle = \langle\langle a_{k\sigma}; a_{k\sigma}^{+}\rangle\rangle. \tag{5}$$

The electronic energy in the generalised Hartree–Fock approximation below T_C is

$$\omega(\boldsymbol{k}, \sigma) = \varepsilon_k - \mu - \sigma(\mu_B H + 0.5I\langle S^z\rangle) + \sum_{k',\sigma'} [v(0) - v(\boldsymbol{k} - \boldsymbol{k}')\delta_{\sigma\sigma'}]\langle m_{k'\sigma'}\rangle \qquad \sigma = \pm 1. \tag{6}$$

$\langle S^z\rangle$ is the localised-spin magnetisation in the direction of the mean field, calculated in I. ε_k is the conduction band energy in the paramagnetic state which for a simple cubic lattice and next-neighbour interaction is given by

$$\varepsilon_k = -\tfrac{1}{3}W(\cos k_x + \cos k_y + \cos k_z) \tag{7}$$

where W is the conduction band width. We do not discuss the results of the electronic energy, the spin magnetisation and the electron polarisation in detail since this was done earlier by other workers (Wesselinowa 1984, 1987, Nolting *et al* 1985, Nolting and Dubil 1985). We listed them in preparation to the discussion of the electronic damping.

4. Dynamic properties

The properties of the 4f and 5f systems are strong functions of the ratio I/W, and we shall see this in the case of the electronic damping γ_{el}^{σ} which was obtained in I to be

$$\begin{aligned}
\gamma_{el}^{+}(\boldsymbol{k}) = {} & \left(\frac{\pi I^2}{4N}\sum_q \bar{n}_{q-k} + \frac{\pi I^2\langle S^z\rangle}{2N}\sum_q \bar{m}_{q-}\right)\delta(E_{q-k} - \omega_{q-} + \omega_{k+}) \\
& + \frac{\pi}{N^2}\sum_{q,p} v_{kqp}^2[\bar{m}_{p-}(1 - \bar{m}_{k-q+} - \bar{m}_{p+q-}) + \bar{m}_{k-q+}\bar{m}_{p+q-}]\delta(\omega_{p-} \\
& - \omega_{p+q-} - \omega_{k-q+} + \omega_{k+}) + \left(\frac{\pi I^2\langle S^z\rangle}{2N^2}\sum_{q,p}(\bar{n}_{p+q-k} + \bar{n}_p)(1 - \bar{m}_{q+})\right. \\
& \left. + \frac{\pi I^2}{4N^2}\sum_{q,p}\bar{n}_{p+q-k}(2\langle S^z\rangle + \bar{n}_p)\right)\delta(E_p - E_{p+q-k} - \omega_{q+} + \omega_{k+})
\end{aligned} \tag{8}$$

$$\begin{aligned}
\gamma_{el}^{-}(\boldsymbol{k}) = {} & \left(\frac{\pi I^2}{4N}\sum_q (\bar{n}_{q-k} + 2\langle S^z\rangle) - \frac{\pi I^2\langle S^z\rangle}{2N}\sum_q \bar{m}_{q+}\right)\delta(-E_{q-k} - \omega_{q+} + \omega_{k-}) \\
& + \frac{\pi}{N^2}\sum_{q,p} v_{kqp}^2[\bar{m}_{p+}(1 - \bar{m}_{k-q-} - \bar{m}_{p+q+}) + \bar{m}_{k-q-}\bar{m}_{p+q+}]\delta(\omega_{p+} \\
& - \omega_{p+q+} - \omega_{k-q-} + \omega_{k-}) + \left(\frac{\pi I^2\langle S^z\rangle}{2N^2}\sum_{q,p}(\bar{n}_{p+q-k} + \bar{n}_p)(1 - \bar{m}_{q-})\right.
\end{aligned}$$

$$+\frac{\pi I^2}{4N^2}\sum_{q,p}\bar{n}_{p+q-k}(2\langle S^z\rangle+\bar{n}_p)\Big)\,\delta(E_p-E_{p+q-k}-\omega_{q-}+\omega_{k-}) \tag{9}$$

with

$$v_{kqp}=(v_q+v_{k-q-p})-(v_{k-q}+v_{p+q})$$
$$\bar{n}_q\equiv\langle S_q^+S_q^-\rangle=(\langle S^z\rangle/2)[(I_\rho/E_q)\coth(E_q/2k_BT)-1]$$
$$\bar{m}_{q\sigma}\equiv\langle a_{q\sigma}^+a_{q\sigma}\rangle=[\exp(\omega_{q\sigma}/k_BT)+1]^{-1}$$

where $\omega_{q\sigma}$ is from (6), E_q and ρ are the spin-wave energy in the generalised Hartree–Fock approximation and the conduction electron magnetisation, respectively (see I). In fact the imaginary parts of the energies corresponding to lifetime-broadening effects are usually not considered because of the mathematical difficulties in their calculations.

The term proportional to $\delta(\omega_{q-}-E_{q-k}-\omega_{k+})$ in (8) represents the scattering of an electronic quasi-particle accompanied by the absorption of a spin wave, whereas the term proportional to $\delta(\omega_{q+}+E_{q-k}-\omega_{k-})$ in (9) represents electronic scattering processes in which a spin wave is emitted. The second terms in (8) and (9) describe scattering processes between electronic quasi-particles due to the Coulomb interaction whereby two electronic quasi-particles result. The last terms in these equations represent scattering processes due to the s–f (or d–f) interaction.

Comparing (8) and (9), we can see that at $T=0$ the electrons with $\sigma=+1$ (in the direction of the spontaneous momentum) are undamped, whereas the electrons with $\sigma=-1$ are damped:

$$\gamma_{el}^-(T=0)=(\pi I^2/2)\langle S^z\rangle \tag{10}$$

provided that the first δ-function can be satisfied. From a physical viewpoint this is understandable, taking into account that in the first case ($\sigma=+1$) the electron can only absorb a spin wave (and at $T=0$ the spin wave does not exist), whereas in the second case ($\sigma=-1$) the electron can only emit a spin wave as a consequence of the total spin conservation of the system with s–d (or d–f) interaction. It should be noted that the first δ-function in (9) can be fulfilled at $T=0$ only for $W>0.5IS=0.35$ eV.

γ_{el}^σ was calculated numerically using the following model parameters: $J_0=0.0001$ eV, $I=\pm0.2$ eV, $S=\frac{7}{2}$, $U\equiv0.5v(\boldsymbol{q})=2$ eV, $\boldsymbol{k}=0$, $H=0$, which should be quite realistic for EuO ($I>0$, $T_C=69.33$ K (Nolting 1979)) and UTe ($I<0$, $T_C=102$ K (Vogt 1980)). Figure 1 ($I>0$) and figure 2 ($I<0$) show the dependence of γ_{el}^- on the ratio $|I/W|$.

Three different regions can clearly be seen (especially for $I<0$; for $I>0$ the intermediate-coupling region is broader—from 0.2 to 0.4):

$$|I/W|\begin{cases}<0.1 & \text{(weak coupling)}\\ =0.1\text{–}(0.2\text{–}0.25) & \text{(intermediate coupling)}\\ >0.3 & \text{(strong coupling).}\end{cases} \tag{11}$$

Typical magnetic semiconductors (e.g. Eu chalcogenides) which belong to the 4f systems are spread throughout the three regions. The Bloch band width W can be estimated from the measured red shift of the optical absorption edge (Nolting *et al* 1985): $W\simeq2.00$ eV (for EuO); 0.90 eV (for EuS); 0.55 eV (for EuSe); 0.45 eV (for EuTe). The s–f coupling constant I is well established to be about 0.2 eV. EuO with $I/W=0.1$ can be assigned to the weak-coupling region and EuS with $I/W=0.22$ to

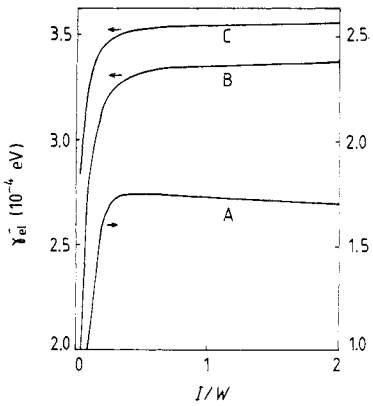

Figure 1. The dependence of the electronic damping γ_{el}^{-} on the ratio I/W for $I = 0.2$ eV, $U = 2$ eV and various temperatures: curve A, $T/T_C = 0.2$; curve B, $T/T_C = 0.6$; curve C, $T/T_C = 0.9$.

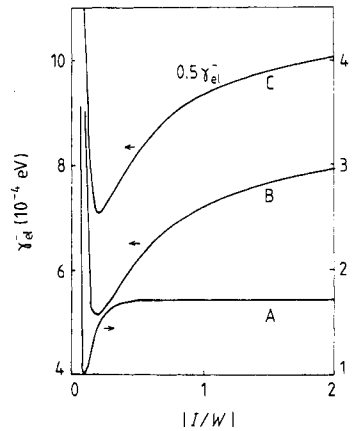

Figure 2. The dependence of the electronic damping γ_{el}^{-} on the ratio $|I/W|$ for $I = -0.2$ eV, $U = 2$ eV and various temperatures: curve A, $T/T_C = 0.2$; curve B, $T/T_C = 0.6$; curve C, $T/T_C = 0.9$.

the intermediate-coupling region, while EuSe and EuTe with $I/W = 0.364$ and 0.444 belong to the strong-coupling region.

Analogous values can be given for the FM U chalcogenides which belong to the 5f systems (Brooks and Glötzel 1980); US and USe with $|I/W| = 0.18$ and 0.25 can be assigned to the weak- and to the intermediate-coupling region, respectively, whereas UTe with $|I/W| = 0.5$ belongs to the strong-coupling region.

In the FM case ($I > 0$) (figure 1) the damping increases very strongly for small I/W-values (weak coupling) and then remains nearly independent of I/W (strong coupling) at high temperatures. At low temperatures, it has a small maximum in the intermediate-coupling region.

The dependence of γ_{el}^{-} on $|I/W|$ in the AFM case ($I < 0$) (figure 2) is quite different. γ_{el}^{-} decreases very rapidly for small $|I/W|$-values (weak coupling). It shows a sharp minimum in the intermediate-coupling region. Then the damping remains nearly independent of $|I/W|$ (strong coupling) at low temperatures or increases strongly at high temperatures. The minimum shifts with increasing temperature from $|I/W| = 0.1$ towards 0.2. Nolting *et al* (1985) reported in the FM case ($I > 0$) a certain critical value $(I/W)_c$ which equals 0.125.

The transition region between atomic-like ($|I/W| \gg 1$) and band-like ($|I/W| \ll 1$) behaviour can be estimated to be in the range $|I/W| = 0.1$–(0.2–0.25) (figure 2). In the FM case this region is broader—from 0.2 to 0.4 (figure 1).

It should be noted that, if the band width W = constant and I increases, then γ_{el}^{-} increases too, and the curve has no minimum.

Figure 3 and figure 4 show the dependence of the electronic damping γ_{el}^{-} on I/W for $U = 0$. The curves have neither a minimum nor a maximum. In the FM case, γ_{el}^{-} increases whereas, in the AFM case, γ_{el}^{-} decreases with increasing $|I/W|$. At large $|I/W|$-values the curves are nearly independent of $|I/W|$. It may be concluded that the Coulomb interaction U plays an important role and must be taken into account if we want to obtain correct results. It will be of interest to calculate the dynamic structure factor via the imaginary part of the Green function. Fuller investigations of the

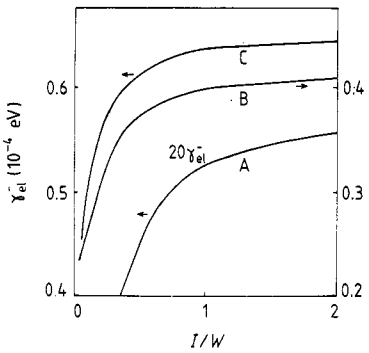

Figure 3. The dependence of the electronic damping γ_{el}^{-} on the ratio I/W for $I = 0.2$ eV, $U = 0$ eV and various temperatures: curve A, $T/T_C = 0.2$; curve B, $T/T_C = 0.6$; curve C, $T/T_C = 0.9$.

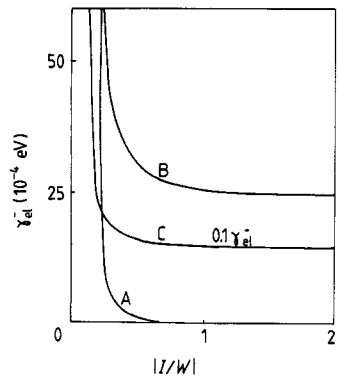

Figure 4. The dependence of the electronic damping γ_{el}^{-} on the ratio $|I/W|$ for $I = -0.2$ eV, $U = 0$ eV and various temperatures: curve A, $T/T_C = 0.2$; curve B, $T/T_C = 0.6$; curve C, $T/T_C = 0.9$.

spectrum including a discussion of the peak height, width and position for different parameters are in preparation and will be published elsewhere.

5. Conclusions

The results presented in this paper surely make clear that the physical properties of magnetic 4f and 5f compounds, and in particular the electronic damping γ_{el}^{-} which is connected to the relaxation time, strongly depend on different parameters such as the s–f (or d–f) coupling constant I, the Bloch band width W, the Coulomb interaction U and the temperature T. Changing the sign of I leads to quite a new physical situation. To our knowledge, the results on $\gamma_{el}^{-}(T, U, |I/W|)$ for $I > 0$ and $I < 0$ are given for the first time.

References

Auslender M I and Bebenin N G 1988 *Fiz. Tverd. Tela* **30** 945
Brooks M S S and Glötzel D 1980 *Physica* B **102** 51
Erbudak M and Meier F 1980 *Physica* B **102** 134
Karmakar S N, Roy G K and Chaudhuri P 1986 *Phys. Lett.* **114A** 145
Nolting W 1979 *Phys. Status Solidi* b **96** 11
Nolting W and Dubil U 1985 *Phys. Status Solidi* b **130** 561
Nolting W, Dubil U and Matlak M 1985 *J. Phys. C: Solid State Phys.* **18** 3687
Reim W 1986 *J. Magn. Magn. Mater.* **58** 1
Shastry B S and Mattis D C 1981 *Phys. Rev.* B **24** 5340
Tserkovnikov Yu A 1971 *Teor. Mater. Fiz.* **7** 250
Vogt O 1980 *Physica* B **102** 206
Wesselinowa J M 1984 *J. Phys. C: Solid State Phys.* **17** 3737
—— 1987 *J. Phys. C: Solid State Phys.* **20** L7