Materials Science-Poland, Vol. 24, No. 3, 2006

Electronic structures of f-electron intermetallic compounds studied by positrons

M. SAMSEL-CZEKAŁA[1*], G. KONTRYM-SZNAJD[1], M. BIASINI[2, 3]

[1]W. Trzebiatowski Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 1410, 50-950 Wrocław 2, Poland

[2]ENEA,via don Fiammelli 2, 40129, Bologna, Italy

[3]Department of Physics, University of California at Riverside, Riverside, California 92521, USA

Electronic structures of the isostructural rare-earth intermetallic compounds $TmGa_3$, $ErGa_3$ and $CeIn_3$ have recently been studied by measurements of two-dimensional angular correlations of annihilation radiation (2D ACAR) spectra. On the example of these systems, we show which kind of information can be exploited from such spectra, i.e. from line projections of the electron-positron momentum density $\rho(\mathbf{p})$. By applying various tomographic techniques, we check how these algorithms can affect reconstructed 3D momentum densities and Fermi surfaces (FS). We confirm that for all these systems in the paramagnetic phase the f-electrons are fully localized and their FSs are similar. $TmGa_3$ and $ErGa_3$ exhibit the FS nesting feature, in agreement with their magnetic structure, which is not the case for $CeIn_3$.

Key words: *electronic structure; positron annihilation; intermetallic compounds*

1. Introduction

The isostructural ($AuCu_3$-type) rare-earth compounds $TmGa_3$, $ErGa_3$, and $CeIn_3$ are interesting due to their magnetic properties, and $CeIn_3$ for its intriguing interplay of antiferromagnetism, heavy-fermion behaviour, and superconductivity when under pressure [1]. Their electronic structures have recently been probed by measurements of 2D ACAR spectra, yielding line projections of the electron-positron (e-p) momentum density $\rho(\mathbf{p})$ in the extended zone $\mathbf{p}$ [2, 3].

Previous studies of $TmGa_3$, $ErGa_3$, and $CeIn_3$ (from three, four, and five 2D ACAR spectra, respectively) point out to the similarity of their electronic structures, both Fermi surface (FS) and f-electrons being localized in the paramagnetic phase

*Corresponding author, e-mail: m.samsel@int.pan.wroc.pl

[4, 5]. $TmGa_3$ and $ErGa_3$ exhibit the FS nesting feature that fits to the observed magnetic structure, which does not occur in $CeIn_3$.

In this work, we discuss the results of reconstructing $\rho(\mathbf{p})$ in these systems by employing different reconstruction algorithms: the modified filtered back projection technique (MFBP) [6], the Cormack method (CM) [7], and new lattice harmonics expansion (LHE) [8] – to the same spectra as in Ref. [4]. Next, we perform Lock –Crisp–West (LCW) folding [9], i.e. conversion from the extended **p** to reduced **k** space, in order to map the FS. The reason for such investigations was that in the case of LaB_6 LHE gave a slightly different FS than other reconstruction algorithms [8].

2. Results and discussion

In this paper, we limit our analysis of densities $\rho(\mathbf{p})$ to their isotropic average $\rho_0(p)$. Due to the similarity of the reconstructed densities after using different algorithms, in Fig. 1 we show only $\rho_0(p)$ reconstructed by LHE. In order to compare the results for various metals, we present them in units of p/p_F and normalized to

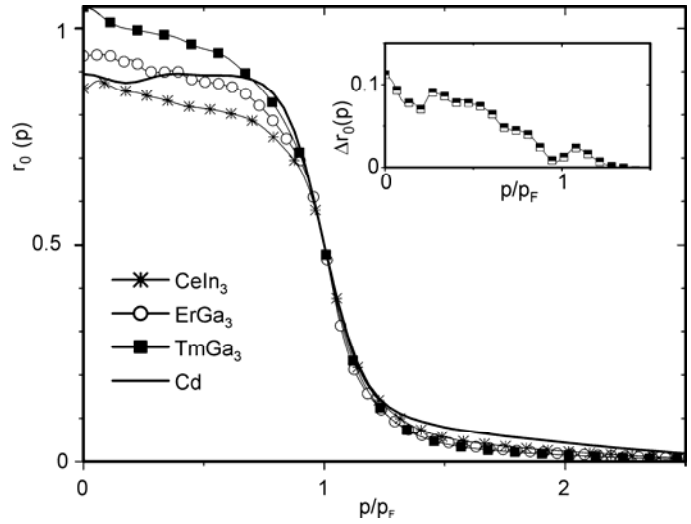

Fig. 1. The isotropic average of momentum densities, $\rho_0(p)$, reconstructed from 2D ACAR spectra of various metals. Inset: the difference between $\rho_0(p)$ in $TmGa_3$ and $ErGa_3$

$\rho_0(p/p_F = 1) = 0.5$. The results for rare-earth compounds are compared with densities reconstructed from 2D ACAR data [10] for Cd for the following reason. According to theoretical calculations [11], $4d^{10}$ electrons in Cd are very close to the conduction band minimum, i.e. they are not as strongly localized as typical core states. It is seen in Fig. 1 (inset) that $\rho_0(p/p_F)$ for rare-earth compounds is lower than for Cd for momenta $p/p_F > 1$. Therefore, knowing that the more localized states there are the

smaller the probability of e-p annihilation (positrons as positive particles are repelled from positive ions), we can conclude that the f-electrons of the considered systems are strongly localized (below the conduction band minimum). A big discrepancy between ρ_0 in these compounds observed for momenta $p/p_F < 1$ comes, most likely, from differences in the compositions of their cores. $TmGa_3$ has twelve 4f electrons per unit cell, i.e. one more than $ErGa_3$. Hence, $\rho_0(p)$ for $TmGa_3$ should be higher than for $ErGa_3$ (as observed), with the difference similar to a typical core contribution (inset of Fig. 1). $CeIn_3$, in turn, has only one 4f electron per unit cell but three atoms of In provide additional (comparing to Ga) $3\times(4s^2+4p^6+4d^{10})$ core electrons. It is therefore somewhat surprising that $\rho_0(p)$ in $CeIn_3$ is smaller than in $ErGa_3$. This indicates that localized f-electrons are relatively close to the bottom of the conduction band minimum.

Obviously, it is impossible to obtain the shape of the FS from the density $\rho(\mathbf{p})$ itself due to the fact that $\rho(\mathbf{p})$ is not constant on the FS and represents a sum of contributions from all occupied bands, not only those crossing the Fermi energy. Thus, in order to map the FS, LCW conversion [9] is usually performed, $\rho(\mathbf{k}) = \Sigma_{\mathbf{G}}\rho(\mathbf{p} = \mathbf{k} + \mathbf{G})$, providing momentum densities in reduced **k** space (**G** are the reciprocal lattice vectors). If the influence of the positron wave function and many-body effects are ignored, $\rho(\mathbf{k})$ depends only on the electron occupation numbers ($n_j(\mathbf{k}) = 0$ or 1) in the j-th band. The total contribution of all bands is then equal to $\Sigma_j n_j(\mathbf{k})=n(\mathbf{k})$, where $n(\mathbf{k})$ denotes the number of occupied bands at the point **k**. In the case of e-p densities, $\rho_j(\mathbf{k}) = n_j(\mathbf{k})g_j(\mathbf{k})$, where the function $g_j(\mathbf{k})$ depends on the electron state $|\mathbf{k}_j>$, even after neglecting e-e and e-p correlations [12,13]. Particularly, small values of g are expected for localized d-like states, because an increase of electron state localization decreases the probability of e-p annihilation. Therefore, if the character of various states $|\mathbf{k}_j>$ varies strongly, their relative contributions to $\rho_j(\mathbf{k})$ may be essentially different. Nevertheless, the values of $g_j(\mathbf{k})$ are usually high enough to reproduce an observable jump in $\rho_j(\mathbf{k})$ if this quantity passes from one band to another [12, 13].

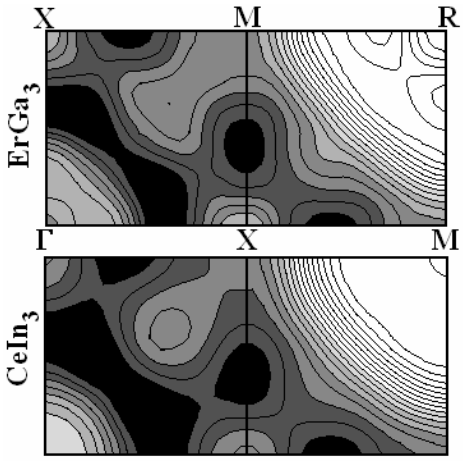

Fig. 2. Densities $\rho(\mathrm{k})$ in $ErGa_3$ and $CeIn_3$ on high symmetry planes, reconstructed using LHE. The white region centred at R contains the occupied states of the 7th valence band, and the dark region corresponds to the unoccupied states of the 6th band

According to theoretical calculations [4] (with f electrons as core states), the FS sheets of $TmGa_3$, $ErGa_3$, and $CeIn_3$ exist in the 6th and 7th valence bands. In Figure 2, we display densities $\rho(\mathbf{k})$ in $ErGa_3$ and $CeIn_3$ on high symmetry planes in the Brillouin zone (BZ), reconstructed by the LHE. It is visible that the general shapes of these densities and thus also of the FSs are similar. The nesting of the FS centred around R (Fig. 2) on the planes perpendicular to the XR line (the [110] direction) is in agreement with the propagation vector of the magnetic structure of $ErGa_3$. If the FS nesting were present in $CeIn_3$, consistent with its antiferromagnetic structure, it would be observed along the [111] direction. No experimental evidence of nesting, however, can be observed. Obviously, even if the electronic structure is in fair agreement with experiment, only a full calculation of Q-dependent magnetic susceptibility can reveal to what extent the adopted band structure calculation is able to account for the magnetic structure of $CeIn_3$.

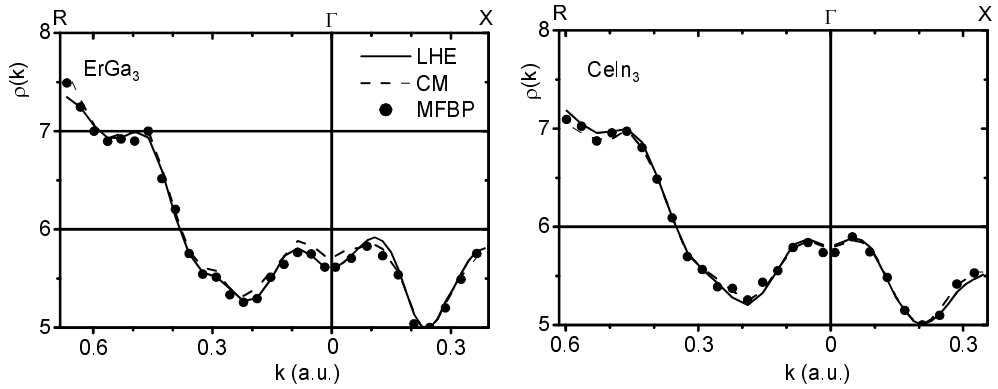

Fig. 3. Densities $\rho(k)$ in $ErGa_3$ and $CeIn_3$ along Γ_X and Γ_R, reconstructed using LHE, CM, and MFBP

In order to show more details, in Fig. 3 we present $\rho(\mathbf{k})$ for $ErGa_3$ and $CeIn_3$ along some high symmetry directions in the BZ, reconstructed by LHE, CM, and MFBP, and normalized to the number of occupied bands in points $\mathbf{k}$. It is seen that $\rho(\mathbf{k})$, very similar for both compounds (the same for $TmGa_3$ not shown here), differs mainly around the R and X points. Around R, where 7 bands are fully occupied, the electron occupation number $n(\mathbf{k})$ is equal to 7. Along the ΓX line and around $k = 0.25$ a.u. (5 bands fully occupied), $n(\mathbf{k}) = 5$ and $n(\mathbf{k}) = 6$ around the Γ and X points. Differences between the occupation numbers $n(\mathbf{k})$ and $\rho(\mathbf{k})$ point out that electrons around R are more free than others and that this effect is more pronounced for $ErGa_3$ than for $CeIn_3$.

3. Conclusions

We presented e-p momentum densities in $TmGa_3$, $ErGa_3$, and $CeIn_3$ as reconstructed by various techniques, showing how a positron annihilation experiment en-

ables the character of electrons to be examined. We confirm that f-electrons in these compounds are strongly localized in the paramagnetic phase and their FSs are similar to one another. The FS nesting feature in $TmGa_3$ and $ErGa_3$ is consistent with the magnetic structure observed by neutrons [14, 15], which is not the case for $CeIn_3$.

Acknowledgements

We are very grateful to the Polish State Committee for Scientific Research (Grant No. 2 P03B 012 25) and the network MAG-EL-MAT in Poznań (Poland) for financial support.

References

[1] GROSCHE F.M., WALKER I.R., JULIAN S.R., MATHUR N.D., FREYE D.M., STEINER M.J., LONZARICH G.G., J. Phys.: Condens. Matter, 13 (2001), 2845 and references therein.
[2] BERKO U.P., [in:] W. Brandt, A. Dupasquier (Eds.), Int. School Phys. Enrico Fermi Course LXXXIII, North-Holland, Amsterdam, 1983, p. 64.
[3] WEST R.N., Adv. Phys., 22 (1973), 263; [in:] A. Dupasquier, A.P. Mills Jr. (Eds.), *Positron Spectroscopy of Solids*, IOS Press, Amsterdam, 1995.
[4] BIASINI M., KONTRYM-SZNAJD G., MONGE M.A., GEMMI M., CZOPNIK A., JURA A., Phys. Rev. Lett., 86 (2001), 4616; BIASINI M., FERRO G., KONTRYM-SZNAJD G., CZOPNIK A., Phys. Rev. B, 66 (2002), 075126; BIASINI M., FERRO G., CZOPNIK A., Phys. Rev. B, 68 (2003), 094513.
[5] RUSZ J., BIASINI M., Phys. Rev. B, 71 (2005), 233103.
[6] BROOKS R.A., DI CHIRO G., Phys. Med. Biol., 21 (1976), 689; KONTRYM-SZNAJD G., JÓZEFCZUK E., Mat. Sci. Forum, 255–257 (1997), 754.
[7] CORMACK A.M., J. Appl. Phys., 34 (1963), 2722; 35 (1964), 2908; KONTRYM-SZNAJD G., Phys. Stat. Sol. (a), 117 (1990), 227.
[8] KONTRYM-SZNAJD G., SAMSEL-CZEKAŁA M., BIASINI M., KUBO Y., Phys. Rev. B, 70 (2004), 125103.
[9] LOCK D.G., CRISP V.H.C., WEST R.N., J. Phys. F: Met. Phys., 3 (1973), 561.
[10] WALTERS P.A., MAYERS J., WEST R.N., [in:] *Positron Annihilation*, Proc. 6th Int. Conference *Positron Annihilation*, The University of Texas at Arlington, April 3–7, 1982, P.G. Coleman, S.C. Sharma, L.M. Diana (Eds.), North-Holland, Amsterdam, 1982, p. 334.
[11] DANIUK S., JARLBORG T., KONTRYM-SZNAJD G., MAJSNEROWSKI J., STACHOWIAK H., J. Phys. Condens. Matter, 1 (1989), 8397.
[12] RABOU L.P.L.M., MIJNARENDS P.E., Solid State Commun., 52 (1984), 933.
[13] KAISER J.H., WEST R.N., SHIOTANI N., J. Phys. F: Met. Phys., 16 (1986), 1307.
[14] MORIN P., GIRAUD M., BURLET P., CZOPNIK A., J. Magn. Magn. Mater., 68 (1987), 107.
[15] MURASIK A., CZOPNIK A., KELLER L., FISCHER P., J. Magn. Magn. Mater., 213 (2000), 101.

Received 1 June 2005