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The relativistic RKKY interaction, uniaxial and unidirectional magnetic anisotropies and spin glasses

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Abstract. We study the interaction between two and three magnetic impurities in a metallic host in which the mediating electrons are modelled by relativistic 'jellium'. We find the relativistic multi-site and multi-component generalisation of the RKKY interaction. In addition to the usual isotropic spin–spin interaction we find uniaxial and unidirectional contributions. We comment on the importance of these to the understanding of magnetic anisotropy of spin glasses.

We shall study the interaction between magnetic impurities in a metallic host. The cases of (i) two and three magnetic impurities and (ii) two magnetic and a single non-magnetic impurity are investigated. The electrons which mediate these interactions will be modelled by an infinite non-interacting electron gas of appropriate density with a uniform, positively charged background (jellium). In that case the conventional coupling between the two magnetic impurities with moments pointing along the unit vectors $\hat{\boldsymbol{s}}_1$ and $\hat{\boldsymbol{s}}_2$ respectively at an asymptotically large separation $\boldsymbol{R}_{12}$, is given by the well known Ruderman–Kittel–Kasuya–Yoshida (RKKY) [1–3] formula.

$$E_{12}^{\mathrm{RKKY}} = V(\hat{\boldsymbol{s}}_1 \cdot \hat{\boldsymbol{s}}_2)[(2k_{\mathrm{F}}R_{12}\cos 2k_{\mathrm{F}}R_{12} - \sin 2k_{\mathrm{F}}R_{12})/R_{12}^4] \tag{1}$$

where k_{F} is the Fermi momentum and V is a parameter that describes the interaction between the impurity and the electrons. The precise nature of V depends on how the impurities are modelled.

As is well known, the above result has been derived within the framework of non-relativistic quantum mechanics. Elsewhere [4] we have used the Dirac equation to describe the conduction electrons and derived the relativistic generalisation of (1). As well as depending on the usual spatially isotropic 'spin–spin' term, on account of spin–orbit coupling effects, it is also a polynomial function of pseudo-dipolar and *squared* Dzyaloshinskii–Moriya type terms. Thus the anisotropy was shown to be uniaxial in form. Here we extend that work to the case of two non-equivalent magnetic impurities and find additional unidirectional terms. We also study the magnetic anisotropic effects arising from three impurity interactions and comment on the more interesting consequences of these results.

While in non-relativistic quantum mechanics the exchange interaction is isotropic in spin space, in the Dirac theory the spin and orbital degrees of freedom are coupled.

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This coupling is the main source of magneto-crystalline anisotropies in metals. Thus the relativistic RKKY interaction is of general interest because it features the simplest example of magnetic anisotropy. Moreover, it is of particular interest because it may make a significant contribution to the still mysterious anisotropies which characterise the low temperature behaviour of the spin glass state [5].

From the point of view of our present concern, the relevant description of the impurities is the corresponding spin-dependent potentials: $\mathbf{v}_i(\boldsymbol{r}) = v_i(\boldsymbol{r})\mathbf{I} + w_i(\boldsymbol{r})\boldsymbol{\sigma}\cdot\hat{\boldsymbol{s}}_i$ where $\mathbf{I}$, $\boldsymbol{\sigma}^x$, $\boldsymbol{\sigma}^y$, and $\boldsymbol{\sigma}^z$ are the complete set of Pauli spin matrices. We wish to work to all orders in $\mathbf{v}_i (i = 1, 2, 3, \ldots)$ and hence we shall develop the theory in terms of the 'on the energy shell' t-matrices

$$\mathbf{t}_i = \left(t^{(i)}_{l,m,\sigma;l^1,m^1,\sigma^1}(\epsilon)\right)$$

where ϵ is the kinetic energy of the incident electron, l, m and σ are the polar, azimuthal and spin magnetic quantum numbers respectively. Note that m and σ refer to a common z axis at both sites and that the matrix $\mathbf{t}(\epsilon)$ is non-diagonal in m and σ. The orbital parts of the potentials are taken to be spherically symmetric and hence the $\mathbf{t}^{(i)}$ are diagonal in l. For orientation we note that in a non-relativistic theory for a local moment along z, $t_{l,m,\sigma;l^1,m^1,\sigma^1}(\epsilon) = -\delta_{l,l^1}\delta_{m,m^1}\delta_{\sigma,\sigma^1}(\exp 2i\delta_{l,\sigma}(\epsilon) - 1)/(2i\sqrt{\epsilon})$. On the other hand in a fully relativistic theory $t_{l,m,\sigma;l^1,m^1,\sigma^1}$ is non-diagonal and its diagonal elements depend on both m and σ. Both of these effects are the consequences of the spin–orbit coupling and in the present framework they are the root causes of magnetic anisotropy. General features of this t-matrix and ways of calculating them have been discussed in [6] and [7].

Under the most general circumstances the quantities that we wish to calculate are the various contributions to the Grand Potential Ω that represent the interaction between 2, 3,... impurities.

$$\Omega = \Omega_0 + \sum_i \delta\Omega_i + \sum_{ij} \delta\Omega_{ij} + \sum_{ijk} \delta\Omega_{ijk} + \cdots. \tag{2}$$

The dominant single particle contribution is given by

$$\Omega = -\int \mathrm{d}\epsilon\ f(\epsilon - \nu) N(\epsilon) \tag{3}$$

where $f(\epsilon - \nu)$ is the Fermi function, ν is the electron chemical potential and N is the appropriate integrated density of states.

A powerful result of multiple scattering theory [7] is that for an arbitrary number and arrangement of non-overlapping scatterers the integrated density of states is given by

$$N(\epsilon) = (1/\pi)\ \mathrm{Im}\log \left\| \begin{matrix} t_1^{-1} & -G_{12} & \cdots \\ -G_{21} & t_2^{-1} & \cdots \\ \vdots & \vdots & \vdots\vdots \end{matrix} \right\| \tag{4}$$

where $\|\boldsymbol{\Theta}\|$ denotes the operation of taking the determinant of the matrix $\boldsymbol{\Theta}$ with respect to the indices l, m, σ and the site indices 1, 2,..., and the matrices $\mathbf{G} \rightarrow$

$(G_{l,m,\sigma;l^1,m^1,\sigma^1}(\boldsymbol{R}_{12};\epsilon))$ are the usual real-space KKR structure constants. They depend on the geometrical arrangement of the scatterers only and are given by

$$G_{l,m,\sigma;l^1,m^1,\sigma^1}(\boldsymbol{R}_{12};\epsilon) = \delta_{\sigma,\sigma^1} 4\pi\sqrt{\epsilon} \sum_{l_2,m_2} \mathrm{i}^{l_2} C^{l_2,m_2}_{l,m;l^1,m^1} Y^{m_2}_{l_2}(\hat{\boldsymbol{R}}_{12}) h^+_{l_2}(\sqrt{\epsilon}R_{12}) \tag{5}$$

where the $Y^m_l(\hat{\boldsymbol{R}})$ are the complex spherical harmonics for the angles which specify the direction $\hat{\boldsymbol{R}}$, the $C^{l_2,m_2}_{l,m;l^1,m^1}$ are the corresponding Gaunt numbers

$$C^{l_2,m_2}_{l,m;l^1,m^1} = \int \mathrm{d}\hat{\boldsymbol{r}}\; Y^m_l(\hat{\boldsymbol{r}}) Y^{m_2\,*}_{l_2}(\hat{\boldsymbol{r}}) Y^{m^1}_{l^1}(\hat{\boldsymbol{r}}) \tag{6}$$

and $h^+_{l_2}$ is the spherical Hankel function for the angular momentum label l_2. For clarity we stress that equation (4) is a straightforward relativistic generalisation of Lloyd's formula [8].

Using equations (4), (2) and (3) we find that

$$\delta\Omega_{12} = -(1/\pi) \int \mathrm{d}\epsilon\; f(\epsilon - \nu) \log |\mathbf{I} - \mathbf{t}_1 \mathbf{G}_{12} \mathbf{t}_2 \mathbf{G}_{21}| \tag{7}$$

where $|\cdots|$ means taking the determinant with respect to the indices l, m and σ only, the operation with respect to the site indices having been fully worked out. A similar analysis provides a more complicated expression for $\delta\Omega_{123}$, the interaction between three sites.

Although the exact formulae of the above form are easy to evaluate numerically, we shall study only their asymptotic forms for large separations between the impurities. In that limit $G_{ij} \sim 1/R_{ij}$ and hence we may expand equation (7) in powers of G_{ij}. For two impurities and to lowest order in G_{ij} we find

$$\delta\Omega_{12} = (1/\pi)\ \mathrm{Im} \int \mathrm{d}\epsilon\; f(\epsilon - \nu) \sum_{\Lambda,\Lambda_1,\Lambda_2,\Lambda_3} \left(t^{(1)}_{\Lambda,\Lambda_1}(\epsilon) G_{\Lambda_1,\Lambda_2}(\boldsymbol{R}_{12},\epsilon) t^{(2)}_{\Lambda_2,\Lambda_3}(\epsilon) G_{\Lambda_3,\Lambda}(\boldsymbol{R}_{21},\epsilon)\right) \tag{8}$$

where Λ denotes all three indices l, m and σ. Similarly, for three impurities

$$\begin{aligned}\delta\Omega_{123} = (1/\pi)\ \mathrm{Im} \int \mathrm{d}\epsilon\; f(\epsilon - \nu) \\ \times \sum_{\Lambda,\Lambda_1,\ldots,\Lambda_6} \big(t^{(1)}_{\Lambda,\Lambda_1}(\epsilon) G_{\Lambda_1,\Lambda_2}(\boldsymbol{R}_{12},\epsilon) t^{(2)}_{\Lambda_2,\Lambda_3}(\epsilon) G_{\Lambda_3,\Lambda_4}(\boldsymbol{R}_{23},\epsilon) t^{(3)}_{\Lambda_4,\Lambda_5}(\epsilon) G_{\Lambda_5,\Lambda}(\boldsymbol{R}_{31},\epsilon) \\ + t^{(1)}_{\Lambda,\Lambda_1}(\epsilon) G_{\Lambda_1,\Lambda_2}(\boldsymbol{R}_{13},\epsilon) t^{(3)}_{\Lambda_2,\Lambda_3}(\epsilon) G_{\Lambda_3,\Lambda_4}(\boldsymbol{R}_{32},\epsilon) t^{(2)}_{\Lambda_4,\Lambda_5}(\epsilon) G_{\Lambda_5,\Lambda}(\boldsymbol{R}_{21},\epsilon)\big).\end{aligned} \tag{9}$$

All explicit calculations which we shall report will have been evaluations of the above formulae by converting the energy integration into a sum over the usual Matsubara frequencies and performing the sum numerically.

To gain some insight into the workings of the formal expression in (8) let us (i) neglect the energy dependence of the t-matrices by replacing both by the real part of $t_{\Lambda,\Lambda^1}(\epsilon_F)$, (ii) use the $T = 0$ K limit of $f(\epsilon - \nu) \approx \Theta(\epsilon - \epsilon_F)$, (iii) take the non-relativistic

limit and (iv) retain only the leading term in $1/R_{12}$. It is then straightforward to show that (8) reduces to

$$\delta\Omega_{12} = \sum_l \{[(t_{l,1/2}(\epsilon_F) + t_{l,-1/2}(\epsilon_F))^2 + (t_{l,1/2}(\epsilon_F) - t_{l,-1/2}(\epsilon_F))^2]\hat{s}_1 \cdot \hat{s}_2\} \times (2k_F R_{12} \cos 2k_F R_{12} - \sin 2k_F R_{12})/R_{12}^4 \tag{10}$$

whose 'magnetic' contribution is the same as in equation (1).

In the non-relativistic derivation it is found that $\delta\Omega_{12}$ depends only on $|\boldsymbol{R}_{12}|$ because $t_{\Lambda,\Lambda'}(\epsilon)$ is diagonal and independent of m. As a consequence of such m-dependence in the relativistic theory a number of new terms containing the 'direction' of $\boldsymbol{R}_{12}$, denoted by the unit vector $\hat{\boldsymbol{R}}_{12}$, arise. Equation (8) may be rewritten, following the same manipulations as in [4], as

$$\delta\Omega_{12} = \sum_{n_1,n_2,n_3}^{2l_{\max}} b_{n_1,n_2,n_3}(\hat{s}_1 \cdot \hat{s}_2) \times [(\hat{\boldsymbol{R}}_{12} \cdot \hat{s}_1)(\hat{\boldsymbol{R}}_{12} \cdot \hat{s}_2)]^{n_1} [\hat{\boldsymbol{R}}_{12} \cdot (\hat{s}_1 \times \hat{s}_2)]^{2n_2} [(\hat{\boldsymbol{R}}_{12} \cdot \hat{s}_1)^2 - (\hat{\boldsymbol{R}}_{12} \cdot \hat{s}_2)^2]^{n_3} \tag{11}$$

where the coefficient $b_{n_1,n_2,n_3}(\hat{s}_1 \cdot \hat{s}_2)$ depends only on the relative orientation of $\hat{s}_1$ and $\hat{s}_2$, R_{12}, the scattering properties of the two impurities and sundry numerical factors. For two identical magnetic impurities $n_3 = 0$ only and $b_{n_1,n_2,0} = a_{n_1,n_2}$ of [4]. This is the general form of the relativistic RKKY interaction.

Evidently, for s wave scattering $l_{\max} = 0$ and hence no new term arises. An example of this case would be two magnetic impurities described by $\mathbf{v}(\boldsymbol{r}) = J\delta(\boldsymbol{r} - \boldsymbol{R})\hat{s}_i \cdot \boldsymbol{\sigma}$ where $\boldsymbol{R}$ is the position of the nucleus. Since we expect pseudo-dipolar terms, like $(\hat{\boldsymbol{R}}_{12} \cdot \hat{s}_1)(\hat{\boldsymbol{R}}_{12} \cdot \hat{s}_2)$, and Dzyaloshinskii–Moriya- (DM-) type terms, like $\hat{\boldsymbol{R}}_{12} \cdot (\hat{s}_1 \times \hat{s}_2)$, to arise only on account of the spin–orbit interaction, this is a satisfactory result as in the case of s states there is no such interaction.

Although equation (11) for two identical impurities contains terms like $\hat{\boldsymbol{R}}_{12} \cdot (\hat{s}_1 \times \hat{s}_2)$ it differs from the usual DM terms in that here only even powers of $\hat{\boldsymbol{R}}_{12} \cdot (\hat{s}_1 \times \hat{s}_2)$ enter. This is a natural consequence of the symmetry in the present problem and hence it is also a very satisfactory feature of our formula. Evidently the magnetic anisotropy described by it is uniaxial. As soon as this interchange symmetry is broken, unidirectional effects occur. This is the case in (11) for two non-equivalent magnetic impurities.Of course, when we consider the three-impurity problem studied by Levy and Fert [5] we also find unidirectional contributions to $\delta\Omega_{123}$ linear in $\hat{\boldsymbol{R}}_{12} \cdot (\hat{s}_1 \times \hat{s}_2)$ (as shown in [4]). As we shall indicate presently such magnetic anisotropic effects are comparable, in magnitude, to those predicted by (11).

Elsewhere [4] we illustrated the consequences of the relativistic RKKY interaction of (11) for two spin-polarised Fe and Co atoms respectively using the corresponding potential functions given in [9]. We compared the interaction energies $\delta\Omega_{12}^{\uparrow\uparrow}$ for the two moments parallel to each other and to $\hat{\boldsymbol{R}}_{12}$ and perpendicular, $\delta\Omega_{12}^{\rightarrow\rightarrow}$, to $\hat{\boldsymbol{R}}_{12}$ for Fe and Co respectively. As expected the anisotropy energy $\delta\Omega_{12}^{\uparrow\uparrow} - \delta\Omega_{12}^{\rightarrow\rightarrow}$ (on the scale of 10^{-6} au) was smaller than the interaction energy $\delta\Omega_{12}^{\uparrow\uparrow}$ by a factor of 10^3. Moreover it was in the range of experimental observation. Interestingly, the anisotropic contributions to the interaction energy oscillates and is of long range like the non-relativistic RKKY interaction. This implies anisotropic frustration in spin glasses [10]. These contributions are uniaxial in form, however, and consequently unable to explain the purely directional

Table 1. The energies of interaction (in mRyd) between a Fe and a Co atom separated by $R_{12} = 3.0\pi$, $k_F = 0.86$ au. Orientational arrangements are shown by the sites 1 and 2 of figure 1.

Configuration	$\delta\Omega_{12}$
I	2.8050
II	2.8054
III	2.8068
IV	2.8068
V	2.8068

character of the magnetic anisotropy of such systems as demonstrated by several types of experiment. These include measurements of shifted hysteresis cycles, NMR and torque measurements [11].

The simplest unidirectional anisotropy occurs when we consider the interaction between two non-equivalent magnetic impurities. This is exemplified by the results shown in table 1, which describe the interaction between an iron and a cobalt impurity separated by a distance 3π au ($k_F = 0.86$ au) with moments whose orientations are specified by the sites 1 and 2 of the five arrangements shown in figure 1. Whereas, for two identical impurities, the interaction of configuration (I) only would differ from the other four, for this new situation, both arrangements (I) and (II) differ from the remaining three. The difference between (II) and (III) in particular demonstrates a unidirectional anisotropy. These interactions again have an oscillatory long ranged dependence on separation.

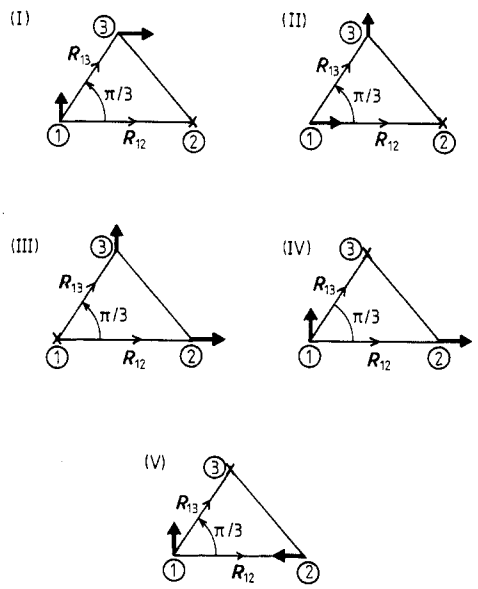

Figure 1. The five configurations used in the calculations described in the text. The calculations involving two atoms only refer to the arrangements specified by sites 1 and 2 only. × denotes a 'spin' perpendicular to the plane in which both $\boldsymbol{R}_{12}$ and $\boldsymbol{R}_{13}$ lie (into the paper). Arrows describes 'spins' lying in this plane. $\hat{\boldsymbol{R}}_{12} \cdot \hat{\boldsymbol{R}}_{13} = \pi/3$.

We now go on to discuss three-site interactions on the basis of (9). Following the same procedures as in the above cases of two site interactions, we have evaluated (9) for a number of 'spin' configurations. The arrangements used are shown schematically in figure 1. Table 2 shows the corresponding interactions between three iron atoms. Two sets of calculations are given appropriate to two different sets of site spacings. The differences emphasise the strong spatial separation dependence of these interaction energies. The pairwise interactions are an order of magnitude larger than that of the triplets but the anisotropies (differences between energies of different configurations) are of roughly the same size. This 'non- convergence' may indicate that magnetic anisotropy is a many impurity effect. As expected from the formal analysis, the pairwise interactions show only uniaxial anisotropy, whereas the three site effects can pick out anisotropies unidirectional in nature as shown, for example, by the difference between configurations (II) and (III).

Table 2. The energies of interaction (in mRyd) between three Fe impurities positioned so that (*a*) $R_{12} = 3.0\pi$ and $R_{13} = 2.9\pi$, $k_F = 0.86$ au and (*b*) $R_{12} = 2.9\pi$ and $R_{13} = 2.7\pi$. Orientational arrangements are shown in figure 1.

Configuration	$\delta\Omega_{12}$	$\delta\Omega_{23}$	$\delta\Omega_{31}$	$\sum\delta\Omega_{ij}$	$\delta\Omega_{123}$
(*a*)					
I	−1.5106	−1.8448	−2.1315	−5.4869	0.1201
II	−1.5124	−1.8456	−2.1315	−5.4895	0.1178
III	−1.5124	−1.8466	−2.1306	−5.4896	0.1178
IV	−1.5124	−1.8448	−2.1306	−5.4878	0.1178
V	−1.5124	−1.8448	−2.1306	−5.4878	0.1200
(*b*)					
I	−2.1294	−2.4375	−2.4086	−6.9755	0.0731
II	−2.1310	−2.4380	−2.4086	−6.9776	0.0737
III	−2.1310	−2.4387	−2.4085	−6.9782	0.0736
IV	−2.1310	−2.4375	−2.4085	−6.9770	0.0737
V	−2.1310	−2.4375	−2.4085	−6.9770	0.0729

In table 3 we show the interactions between two iron and one platinum impurities. Platinum was chosen owing to its associated large spin–orbit coupling. The first set of results show the various interaction energies. They were obtained by evaluating (9) fully. Note that the two site interactions are much larger than those of purely three site origin. Moreover, the triplet anisotropy is larger than the pairwise anisotropies as well as being unidirectional. Additionally the second and third columns describe a single magnetic impurity anisotropy. The second set of results examine the consequence of neglecting the spin–orbit coupling associated with platinum. The three-site anisotropy is now minute. Finally the third set of results show the effect of neglecting spin–orbit coupling associated with the magnetic iron atoms. The three site anisotropy is now again fairly large though only a fraction of that from the full calculation. In this limit the two-site anisotropies are necessarily zero.

It should be stressed that these calculations have been performed to illustrate the qualitative features of magnetic anisotropies which arise from relativistic electron scattering. The spin-polarised impurity potentials have been taken from band structure calculations and to that extent reflect real properties of the materials in question. The jellium model which we treat, however, was not designed to give a realistic picture of

Table 3. The energies of interaction (in mRyd) between two Fe impurities on sites 1 and 2 and a Pt site on site 3 of figure 1. $R_{12} = 3.0\pi$ and $R_{13} = 2.9\pi$, $k_F = 0.86$ au. (*a*) Full interaction, (*b*) platinum site treated non-relativistically (i.e. no spin–orbit coupling), (*c*) iron sites treated non-relativistically.

Configuration	$\delta\Omega_{FeFe}$	$\delta\Omega_{Fe^2Pt}$	$\delta\Omega_{PtFe^1}$	$\sum\delta\Omega_{ij}$	$\delta\Omega_{FeFePt}$
(*a*)					
I	−1.5106	−0.5490	−0.9490	−3.0086	0.0802
II	−1.5124	−0.5490	−0.9477	−3.0091	0.0803
III	−1.5124	−0.5498	−0.9470	−3.0092	0.0832
IV	−1.5124	−0.5498	−0.9490	−3.0112	0.0786
V	−1.5124	−0.5498	−0.9490	−3.0112	0.0861
(*b*)					
I	−1.5106	−2.2982	−2.6217	−6.4305	0.1151
II	−1.5124	−2.2982	−2.6208	−6.4314	0.1152
III	−1.5124	−2.2987	−2.6204	−6.4315	0.1152
IV	−1.5124	−2.2987	−2.6217	−6.4328	0.1153
V	−1.5124	−2.2987	−2.6217	−6.4328	0.1152
(*c*)					
I	−1.8213	−0.7325	−1.1319	−3.8657	0.0967
II	−1.8213	−0.7325	−1.1319	−3.8657	0.0967
III	−1.8213	−0.7325	−1.1319	−3.8657	0.0967
IV	−1.8213	−0.7325	−1.1319	−3.8657	0.0954
V	−1.8213	−0.7325	−1.1319	−3.8657	0.0979

electrons in actual dilute alloys. Consequently we do not attempt to interpret actual experiments or compare with other calculations [5], [11], [12].

In short we have presented a general formalism for the study of the interaction between impurities in a metallic host modelled by relativistic jellium. A general relativistic multi-site, multi-component RKKY interaction has been found with a transparent functional form for the two-site limit. Specific calculations relevant to two and three impurities have been carried out. Both uniaxial and unidirectional magnetic anisotropic effects appear naturally. It would be illuminating if the full implications arising from the anisotropic form of these interactions were to be explored by computer simulation studies of spin-glass models.

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