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Enhancement of the spin–orbit coupling by strong electronic correlations in transition metal and light actinide compounds

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Abstract

A simple variational argument is presented which indicates that the spin–orbit coupling in itinerant systems can be enhanced by strong electronic correlations. The importance of the enhancement in the formation of the giant magnetic anisotropy found in the metallic paramagnetic and magnetically ordered states of compounds containing transition metal and light actinide elements (such as tetragonal Sr_2RhO_4, Sr_2IrO_4, the cubic uranium monochalcogenides and tetragonal URu_2Si_2) is discussed.

Keywords: magnetic anisotropy, spin orbit coupling, electronic correlations, enhancement

1. Introduction

Spin–orbit coupling has been a subject of fundamental interests for many decades, starting with John Van-Vleck's research on insulating solids [1] and its extension to metals [2]. In metals, treating the electrons as non-interacting within a tight-binding approximation, it was found [2] that the orbital angular momentum is partially quenched, but the spin–orbit coupling to magnetically ordered spins partially revives the orbital magnetization. The subject of spin–orbit coupling is also of great technological importance [3]. The technological importance stems from the central role that permanent magnets play in many devices and their use in energy conversion and power generation [4]. One of the most important properties of permanent magnets is the anisotropy that arises from the spin–orbit interaction. Highly accurate state-of-the-art DFT electronic structure calculations of the magnetic anisotropy of 3d metals, such as bcc Fe, hcp Co, and fcc Ni, yield results that are in disagreement with experiments [5, 6]. These calculations do not even reproduce the correct easy-axis for Ni. This failure has been attributed to the either the omission of the orbital correlations induced by the intra-atomic Coulomb interactions [7, 8] or the inability of density functional theory to calculate energy differences which are as small as 10^{-6} eV per atom. On the other hand, there have been successful predictions of the magnetic anisotropies of magnetic impurities in non-magnetic hosts due to electronic correlations [9]. The inclusion of electronic correlations in LDA + U [10] and LDA + DMFT [11] calculations of the orbital magnetization of 3d transition metals compounds have produced good results.

Recently, the subject of spin–orbit coupling has undergone a major resurgence of interest. The resurgence of interest in this subject is partly caused by the realization that it plays an essential role in spintronics [12] and in topological materials, such as the so-called topological insulators [13] and Weyl semimetals [14–16]. The resurgence is also partly due to the realization that spin–orbit coupling can play an important role in controlling the metal insulator transition in strontium irradiates [17, 18]. Recent studies on the uranium based superconductor UTe_2 [19] have shown that a dramatic change in the magnetic anisotropy occurs at the ferromagnetic instability [20], which has been attributed to strong spin–orbit coupling [21]. It has also be shown [22] that, by using an itinerant LSDA description, spin–orbit coupling describes the giant magnetic anisotropy in the large moment

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anti-ferromagnetic state [23] of URu_2Si_2. This giant anisotropy is not a consequence of spontaneous symmetry breaking, since similarly large anisotropies are also found in the paramagnetic and Hidden Ordered states [24–27]. However, RPA + LDA calculations of the magnetic anisotropy of the paramagnetic susceptibility of URu_2Si_2 [28], utilizing the atomic spin–orbit splitting, produces an anisotropy of about 1.3 which is significantly smaller than the experimentally observed value of about 10 [24, 29].

In this note we extend the hypothesis of Jansen [7, 8], Shishidou *et al* [10] and Zhu *et al* [11] to include paramagnetic metals [9] and indicate that giant magnetic anisotropy may be generated by strong Coulomb and Hund's rule correlations in the paramagnetic states of metallic compounds containing transition metal and light actinide elements. Specifically, the manuscript illustrates the effects that electronic correlations have on the apparent magnitude of spin–orbit coupling in paramagnetic metals [9, 30].

An enhancement of the spin–orbit coupling may be expected to induce extremely large changes in the magnetic anisotropy. Qualitatively, the origin of the magnetic anisotropy in tetragonal materials can be viewed as being due to the strong coupling between orbital moments to the crystalline structure that hinders their re-orientation due to weak applied magnetic fields. On the other hand, the spins are free to rotate and align with the applied field, so the spin–orbit coupling can be viewed as providing a strong effective magnetic field which can reorient the orbital moment [31]. Specifically, if one considers non-interacting electrons and treat the spin–orbit coupling strength, λ as a perturbation, then up to second-order the anisotropy energy, ΔE, can be expressed in terms of the spin moments S^α as

$$\Delta E = \sum_{\alpha,\beta} \chi^{\alpha,\beta}_{L,L} \left[\lambda^2 S^\alpha S^\beta + 2\mu_B \lambda S^\alpha B^\beta + \mu_B^2 B^\alpha B^\beta\right] \quad (1)$$

where $\chi^{\alpha,\beta}_{L,L}$ is the orbital susceptibility

$$\chi^{\alpha,\beta}_{L,L} = \sum_{n,m} \rho_n \frac{< n|\hat{L}^\alpha|m >< m|\hat{L}^\beta|n >}{E_m - E_n} \quad (2)$$

and $\hat{L}^\alpha$ denotes the components of the orbital angular momentum operator and $\hat{\rho}_n$ is the statistical operator. Hence, the dominant contribution to the magnetic anisotropy is expected to scale with the square of the spin–orbit coupling strength. For cubic materials, the orbital susceptibility exhibits cubic symmetry, so one expects that the magnetic anisotropy will vanish to this order of perturbation approximation and only be found in fourth or higher-order perturbation approximations [31]. Thus, since the magnetic anisotropy is expected to be second-order in the spin–orbit interaction in tetragonal materials and fourth-order in cubic materials, even a modest enhancement of the spin–orbit coupling may produce an extremely large anisotropy.

In the next section, we shall introduce a model Hamiltonian appropriate to describe itinerant $5d$ and $5f$ electron systems. In the following section, we shall introduce the variational approximation. This will be followed by the description of how, in the absence of spin–orbit coupling, electronic correlations can enhance the spin–orbit interaction. The final section discusses and summarizes the results.

2. The Hamiltonian

The on-site Coulomb and Hund's rule interaction between electrons in an atomic shell with angular momentum $l = 3$, can be written as [32–34]

$$\begin{aligned}\hat{H}_{\rm int} = &\frac{1}{2}\sum_{i,m,m',\sigma} U \hat{n}_{i,m,\sigma}\hat{n}_{i,m',\overline{\sigma}} + \frac{1}{2}\sum_{i,m\neq m',\sigma}(U-J)\hat{n}_{i,m,\sigma}\hat{n}_{i,m',\sigma} \\ &- \frac{1}{2}\sum_{i,m\neq m',\sigma} J f^\dagger_{i,m,\sigma} f_{i,m,\overline{\sigma}} f^\dagger_{i,m',\overline{\sigma}} f_{i,m',\sigma} \\ &+ \frac{1}{2}\sum_{i,m,\sigma} J \hat{n}_{i,m,\sigma}\hat{n}_{i,m,\overline{\sigma}},\end{aligned} \quad (3)$$

where $f^\dagger_{i,m,\sigma}$ and $f_{i,m,\sigma}$ create and annihilate an electron in an atomic f orbital localized at site i, with the z-component of the orbital angular momentum m and spin σ. The quantities $U_{m,m'}$ and $J_{m,m'}$ are, respectively, the Coulomb and the Hund's rule exchange interactions which should be dependent on m and m' and are related to the Slater integrals [35]. However, following Anderson [32], we shall assume that the interaction strengths are independent of m and m'. The quantity U represents the strength of the inter-orbital direct Coulomb interaction. The quantity J represents the exchange interaction between f electrons with parallel spins. To be spin-rotationally invariant, the exchange interaction should have the form of a scalar product

$$-J\sum_{m,m'} \underline{\hat{S}}_m . \underline{\hat{S}}_{m'}. \quad (4)$$

The z-component of the scalar product is included in the second line, while the spin-flip components are included are shown in the third line. The last term in equation (3) is necessary for invariance under orbital rotations to be preserved. The requirement of invariance under orbital rotations, leads to the identification of $U' = U + J$ as the direct intra-orbital Coulomb interaction. This differs from the commonly used relation $U' = U + 2J$ since, in the atomic representation, the pair hopping terms are identically zero as they do not conserve the z-component of the total orbital angular momentum. The interaction has $SU(2) \times SO(3)$ symmetry but we describe the $SO(3)$ rotation group by a $(2l + 1)$-dimensional representation with a basis that is composed of the spherical harmonics[6]. The interaction is combined with a Hamiltonian describing the

[6] The mapping of $SU(2)$ onto $SO(3)$ is two to one and cannot simply be considered a representation of $SO(3)$. However, the introduction of a projective or ray-representation [36] with a topologically determined phase factor allows one to treat the mapping $SU(2) \to SO(3)$ as a representation of $SO(3)$. Alternatively, following the suggestion of Bethe [37], one may extend the range of rotations by introducing an enigmatic operator that produces rotations through 2π which is not equivalent to the identity. The addition of this enigmatic quasi-identity operator to the group of rotation operators of $I\,R^3$ doubles the order of the group. The generalized or doubled $SO(3)$ matrix group allows a one to one mapping between it and $SU(2)$.

Bloch bands of non-relativistic non-interacting electrons

$$\sum_{\underline{k},\alpha,\sigma} \epsilon^0_\alpha(\underline{k}) f^\dagger_{\underline{k},\alpha,\sigma} f_{\underline{k},\alpha,\sigma} \tag{5}$$

where $\epsilon^0_\alpha(\underline{k})$ is the band energy of the αth Bloch band with the Bloch wave vector $\underline{k}$. Since the Coulomb interactions are spin-rotationally invariant, the spin σ is a good quantum number in the absence of spin–orbit coupling. The atomic spin–orbit coupling interaction can be expressed as

$$\begin{aligned}\hat{H}_{\rm SO} = \frac{\lambda}{2}\sum_{i,m,m',\sigma} &< m,\sigma|L^z\sigma^z|m',\sigma > f^\dagger_{i,m,\sigma} f_{i,m',\sigma} \\ &+ \frac{\lambda}{2}\sum_{i,m,m'}\Big[< m,\downarrow|\hat{L}^+\hat{S}^-|m',\uparrow > f^\dagger_{i,m,\downarrow} f_{i,m',\uparrow} \\ &+ < m,\uparrow|\hat{L}^-\hat{S}^+|m',\downarrow > f^\dagger_{i,m,\uparrow} f_{i,m',\downarrow}\Big]\,. \end{aligned} \tag{6}$$

We shall neglect the effect that the spin–orbit interaction generated at one atomic site has on a neighbouring atomic site since we argue that this is small. The argument is based on the fact that the non-local spin–orbit coupling falls off proportional to r^{-3} where r is the distance between the two sites and the fact that r is large compared to the linear dimension of the atomic wave functions.

3. The variational approximation

One method of treating many-body correlations is to use the equations of motion. The equations of motion for fermionic operators lead to a set of coupled equations that involve higher-order fermionic operators. In general, the set of coupled equations do not terminate. A commonly used truncation scheme simply approximates higher-order fermionic operators by introducing expectation values. Roth has proposed that, instead of replacing factors in the higher-order fermionic operators by their expectation values, one should evaluate their commutation relations with the other members of the set of fermionic operators and then take the thermal average [38]. This process can be used to restrict the set of operators to a set for which the expectation values are positive definite and the resulting energy eigenvalues are real. As Roth [38–40] noticed, the resulting eigenvalue equation suggests that there is a connection with a variational principle. Sawada [41], using the Gibb's–Bogoliubov inequality, showed that the truncation scheme restricts the operators to those which span a sub-space of Hilbert space, thereby establishing the variational nature of Roth's hierarchy of truncation schemes. Roth's method is illustrated in the first sub-section of the appendix . In the second subsection of the appendix , we shall outline the method of derivation of the mean-field form of the Coulomb interaction shown in equation (3) when the set of operators are truncated to single-electron operators and then display the forms of the various terms.

In this section, we shall display the form of the mean-field Hamiltonian for a system which is totally spin-rotationally invariant. The diagonal and off-diagonal single-electron expectation values are denoted by

$$\begin{aligned}\overline{n}_{i,m,\sigma} &= < f^\dagger_{i,m,\sigma} f_{i,m,\sigma} > \\ \Phi_{i;m,\sigma;m',\sigma'} &= < f^\dagger_{i,m,\sigma} f_{i,m',\sigma'} > \end{aligned} \tag{7}$$

and are to be considered as independent variational parameters. In the absence of spin–orbit coupling, in a paramagnetic state and in the absence of magnetic fields, the system is spin-rotationally invariant. In this case, the mean-field interaction reduces to [9, 42]

$$\begin{aligned}\hat{H}^{\rm MF}_{\rm int} = &\frac{1}{2}\sum_{i,m,m',\sigma} U(\hat{n}_{i,m,\sigma}\overline{n}_{i,m',\overline{\sigma}} + \overline{n}_{i,m,\sigma}\hat{n}_{i,m',\overline{\sigma}}) \\ &- \frac{1}{2}\sum_{i,m,m',\sigma} U\overline{n}_{i,m,\sigma}\overline{n}_{i,m',\overline{\sigma}} + \frac{1}{2}\sum_{i,m\neq m',\sigma}(U-J)(\hat{n}_{i,m,\sigma}\overline{n}_{i,m',\sigma} \\ &+ \overline{n}_{i,m,\sigma}\hat{n}_{i,m',\sigma}) - \frac{1}{2}\sum_{i,m\neq m',\sigma}(U-J)\overline{n}_{i,m,\sigma}\overline{n}_{i,m',\sigma} \\ &+ \frac{1}{2}\sum_{i,m,\sigma} J(\hat{n}_{i,m,\sigma}\overline{n}_{i,m,\overline{\sigma}} + \overline{n}_{i,m,\sigma}\hat{n}_{i,m,\overline{\sigma}}) - \frac{1}{2}\sum_{i,m,\sigma} J\overline{n}_{i,m,\sigma}\overline{n}_{i,m,\overline{\sigma}} \\ &- \frac{1}{2}\sum_{i,m\neq m',\sigma}(U-J) \\ &\times\left(f^\dagger_{i,m,\sigma} f_{i,m',\sigma}\Phi_{i;m',\sigma;m,\sigma} + \Phi_{i;m,\sigma;m',\sigma} f^\dagger_{i,m',\sigma} f_{i,m,\sigma}\right) \\ &+ \frac{1}{2}\sum_{i,m\neq m',\sigma}(U-J)\Phi_{i;m,\sigma;m',\sigma}\Phi_{i;m',\sigma;m,\sigma} \\ &+ \frac{1}{2}\sum_{i,m\neq m',\sigma} J \\ &\times\left(f^\dagger_{i,m,\sigma} f_{i,m',\sigma}\Phi_{i;m',\overline{\sigma};m,\overline{\sigma}} + \Phi_{i;m,\sigma;m',\sigma} f^\dagger_{i,m',\overline{\sigma}} f_{i,m,\overline{\sigma}}\right) \\ &- \frac{1}{2}\sum_{i,m\neq m',\sigma} J\Phi_{i;m,\sigma;m',\sigma}\Phi_{i;m',\overline{\sigma};m,\overline{\sigma}}. \end{aligned} \tag{8}$$

As the system is spin-rotationally invariant, the correlations between opposite spins vanish

$$\Phi_{i;m,\sigma;m',\overline{\sigma}} = 0 \tag{9}$$

and have been omitted. A derivation of a typical term of the mean-field interaction energy is given in equation (39) through (47) of the appendix . In what follows, we shall consider the uniform state in which the expectation values are independent of the site index i. Since the mean-field Hamiltonian is quadratic in the fermion operators it can always be diagonalized leading to renormalized quasiparticle dispersion relations $\epsilon_{\alpha,\sigma}(\underline{k})$ that are described by a new set of quantum numbers (α,σ). Thus, following the work of Sawada [41], the Gibbs–Bogoliubov variational grand-canonical free-energy, $\Omega_{\rm V}$, is calculated as

$$\begin{aligned}\Omega_{\rm V} = &\sum_{\underline{k},\alpha,\sigma}(\epsilon_{\alpha,\sigma}(\underline{k}) - \mu) f_{\alpha,\sigma}(\underline{k}) + k_B T\sum_{\underline{k},\alpha,\sigma} \\ &\times\left[f_{\alpha,\sigma}(\underline{k})\ln f_{\alpha,\sigma}(\underline{k}) + (1 - f_{\alpha,\sigma}(\underline{k}))\ln(1 - f_{\alpha,\sigma}(\underline{k}))\right]\end{aligned}$$

$$- \frac{N}{2}\left[\sum_{m,m',\sigma} U\overline{n}_{m,\sigma}\overline{n}_{m',\overline{\sigma}} + \sum_{m\neq m',\sigma}(U-J)\overline{n}_{m,\sigma}\overline{n}_{m',\sigma} + \sum_{m,\sigma} J\overline{n}_{m,\sigma}\overline{n}_{m,\overline{\sigma}}\right] + \frac{N}{2}\sum_{m\neq m',\sigma} \times \left[(U-J)\Phi_{m,\sigma;m',\sigma}\Phi_{m',\sigma;m,\sigma} - J\Phi_{m,\sigma;m',\sigma}\Phi_{m',\overline{\sigma};m,\overline{\sigma}}\right]. \tag{10}$$

The first line represents the quasiparticle energy contribution to the free-energy, $E_{\rm qp} - \mu N$. The second line describes the entropy. In these lines, $f_{\alpha,\sigma}(\underline{k})$ is considered to be a quantum statistical quasiparticle distribution function that is to be determined variationally. The last line displays corrections that avoid double-counting the interaction energy. Minimization of $\Omega_{\rm V}$ w.r.t. $f_\alpha(\underline{k})$ results in $f_\alpha(\underline{k})$ being identified with the Fermi-function. Minimization of $\Omega_{\rm V}$ w.r.t the expectation values $\overline{n}_{m,\sigma}$, leads to a set of equations

$$\frac{1}{N}\sum_{\underline{k},\alpha,\sigma'}\left(\frac{\partial\epsilon_{\alpha,\sigma'}(\underline{k})}{\partial\overline{n}_{m,\sigma}}\right) f_{\alpha,\sigma'}(\underline{k}) = U\sum_{m'}\overline{n}_{m',\overline{\sigma}} + (U-J)\sum_{m'\neq m}\overline{n}_{m',\sigma} + J\overline{n}_{m,\overline{\sigma}}. \tag{11}$$

The term on the left-hand side originates with the quasiparticle energy and the terms on the right-hand side come from the double-counting corrections to the total energy. Since $f_{\alpha,\sigma}(\underline{k})$ is an independent variational parameter, the derivative only acts on the quasi-particle energies. The free-energy is minimized when the inter-band correlations satisfy the equations

$$\frac{1}{N}\sum_{\underline{k},\alpha,\sigma'}\left(\frac{\partial\epsilon_{\alpha,\sigma'}(\underline{k})}{\partial\Phi_{m,\sigma;m',\sigma}}\right) f_{\alpha,\sigma'}(\underline{k}) = (U-J)\sum_{m'\neq m}\Phi_{m',\sigma;m,\sigma} + J\Phi_{m',\overline{\sigma};m,\overline{\sigma}}. \tag{12}$$

The above sets of equations have to be solved self-consistently.

In the absence of spin–orbit coupling, the vanishing of $\Phi_{m,\sigma;m',\overline{\sigma}}$ can be demonstrated by minimizing the general expression for $\Omega_{\rm V}$ w.r.t. the conjugate field $\Phi_{m',\overline{\sigma};m,\sigma}$. In the absence of spin–orbit coupling and applied magnetic fields, the left-hand side of the self-consistency equation for $\Phi_{m,\sigma;m',\overline{\sigma}}$, does not contain source terms. Hence in the absence of spin–orbit coupling, the self-consistency equations reduce to a set of homogeneous equations, so finite off-diagonal spin correlations can only be generated by spontaneous symmetry breaking [42].

In the next section, we shall show that the electronic correlations induced by U result in an enhancement of the spin–orbit coupling.

4. Enhanced spin–orbit coupling

In this section, we examine the enhancement of the spin–orbit coupling that is produced by treating the Coulomb interaction U with Roth's method. The one-electron spin–orbit coupling is expressed as the sum of components that are longitudinal and transverse to the axis of quantization

$$\hat{H}_{\rm SO} = \lambda\left[\hat{L}^z\hat{S}^z + \frac{1}{2}(\hat{L}^+\hat{S}^- + \hat{L}^-\hat{S}^+)\right]. \tag{13}$$

First, we describe the enhancement of the transverse components which will be followed by the description of the enhancement of the longitudinal component of the spin–orbit coupling.

In the absence of magnetic order and applied magnetic fields, the only single-electron terms in the Hamiltonian that are off-diagonal in both the spin and orbital quantum numbers are the transverse terms in the spin–orbit coupling. The transverse spin–orbit coupling has the same form as terms displayed in equation (48) of the appendix which are generated by the mean-field approximation to the Coulomb interaction U between electrons with antiparallel spins. Therefore, the spin–orbit coupling and part of the off-diagonal mean-field Coulomb correlations can be combined into an effective spin–orbit coupling $\hat{\tilde{H}}^{\pm}_{\rm SO}$ given by

$$\hat{\tilde{H}}^{\pm}_{\rm SO} = \sum_{i,m}\left[< m,\downarrow|\hat{\tilde{H}}^{+}_{\rm SO}|m-1,\uparrow> f^{\dagger}_{i,m,\downarrow}f_{i,m-1,\uparrow} + < m-1,\uparrow|\hat{\tilde{H}}^{-}_{\rm SO}|m,\downarrow> f^{\dagger}_{i,m-1,\uparrow}f_{i,m,\downarrow}\right], \tag{14}$$

where the interaction matrix elements are given by [9]

$$< m,\downarrow|\hat{\tilde{H}}^{+}_{\rm SO}|m-1,\uparrow> = \left(\frac{\lambda}{2} < m,\downarrow|\hat{L}^+\hat{S}^-|m-1,\uparrow> - U\Phi_{m-1,\uparrow;m,\downarrow}\right) \tag{15}$$

and the Hermitean conjugate expression

$$< m-1,\uparrow|\hat{\tilde{H}}^{-}_{\rm SO}|m,\downarrow> = \left(\frac{\lambda}{2} < m-1,\uparrow|\hat{L}^-\hat{S}^+|m,\downarrow> - U\Phi_{m,\downarrow;m-1,\uparrow}\right). \tag{16}$$

Diagonalization of the resulting mean-field Hamiltonian yields the quasi-particle dispersion relations $\epsilon_\alpha(\underline{k})$ that are functions of $\Phi_{m,\downarrow;m-1,\uparrow}$ and $\overline{n}_{m,\sigma}$. The quasi-particle energies contribute the term

$$E_{\rm qp} = \sum_{\underline{k},\alpha}\epsilon_\alpha(\underline{k})f_\alpha(\underline{k}) \tag{17}$$

to the variational free-energy $\Omega_{\rm V}$. Minimization of $\Omega_{\rm V}$ with respect to the off-diagonal expectation value $\Phi_{m,\downarrow;m-1,\uparrow}$ yields the non-linear relation

$$0 = \frac{1}{N}\sum_{\underline{k},\alpha}\left(\frac{\partial\epsilon_\alpha(\underline{k})}{\partial\Phi_{m,\downarrow;m-1,\uparrow}}\right) f_\alpha(\underline{k}) + U\Phi_{m-1,\uparrow;m,\downarrow} \tag{18}$$

where the first term originates from the quasi-particle energy, $E_{\rm qp}$ and the second term originates from the double-counting

correction. The derivative of the quasi-particle energy w.r.t. spin–orbit correlations can be used to define the relation

$$\frac{1}{N}\sum_{\underline{k},\alpha}\left(\frac{\partial\epsilon_\alpha(\underline{k})}{\partial\Phi_{m,\downarrow;m-1,\uparrow}}\right)f_\alpha(\underline{k}) = U\sum_{m'}\chi^{m'-1,m'}_{m,m-1} < m'-1,\uparrow|\hat{\tilde{H}}^{+}_{\rm SO}|m',\downarrow> . \quad (19)$$

which recognizes that the Bloch states are linear superpositions of states with different values of m. The above equations have the appearance of a linear response relation, however, they are strictly non-linear since the susceptibilities are defined in terms of the derivatives of the band energies $\epsilon_\alpha(\underline{k})$ calculated in the presence of the spin–orbit coupling. The presence of the factor $\hat{\tilde{H}}^{+}_{\rm SO}$ is required by time-reversal invariance and inversion symmetry, as can be seen by considering the weak-coupling limit where linear response theory can be applied. On substituting the above definition in equation (18), one obtains the matrix equation

$$\mathbf{\Phi} = -\chi\tilde{\mathbf{H}}_{\rm SO} = -\chi\left[\mathbf{H}_{\rm SO} - U\mathbf{\Phi}\right]. \quad (20)$$

The matrix equation can be formally inverted to yield an expression for the off-diagonal correlations in terms of the bare spin–orbit interaction strength $H_{\rm SO}$,

$$\mathbf{\Phi} = -[\mathbf{I} - U\chi]^{-1}\chi\,\mathbf{H}_{\rm SO}. \quad (21)$$

This equation serves as a self-consistency equation for the off-diagonal electronic correlations $\mathbf{\Phi}$. The effective strength of the spin–orbit coupling is then found to be given by

$$\tilde{\mathbf{H}}_{\rm SO} = \mathbf{H}_{\rm SO} - U\mathbf{\Phi} = [\mathbf{I} - U\chi]^{-1}\mathbf{H}_{\rm SO}. \quad (22)$$

Thus, the transverse part of the spin–orbit interaction is enhanced by the presence of the Coulomb interaction U.

Similarly, the longitudinal component of the spin–orbit coupling is also enhanced by U. In the presence of spin–orbit coupling, the diagonal variational parameters that minimize the variational function satisfy the equation

$$\frac{1}{N}\sum_{\underline{k},\alpha}\left(\frac{\partial\epsilon_\alpha(\underline{k})}{\partial\overline{n}_{m,\sigma}}\right)f(\epsilon_\alpha(\underline{k})) = U\sum_{m',\sigma'}\overline{n}_{m',\sigma'} - J\sum_{m'}\overline{n}_{m',\sigma} - U\overline{n}_{m,\sigma} + J\sum_{\sigma'}\overline{n}_{m,\sigma'} \quad (23)$$

and a similar equation holds in the absence of the longitudinal component of the spin–orbit coupling. The spin–orbit induced correlations between m and σ are denoted by $\Delta\overline{n}_{m,\sigma}$. Since the induced spin–orbit correlations $\Delta\overline{n}_{m',\sigma'}$ have the same symmetry as the product $m'\times\sigma'$ they vanish when either m' or σ' are summed over. Hence, the induced spin–orbit correlations in the first, second and fourth terms of the right-hand side of the equation vanish. Therefore, the induced longitudinal spin–orbit correlations are given by

$$U\Delta\overline{n}_{m,\sigma} = \frac{1}{N}\sum_{\underline{k},\alpha}\left(\frac{\partial\epsilon_\alpha(\underline{k})}{\partial\overline{n}_{m,\sigma}}\right)f(\epsilon_\alpha(\underline{k}))\Big|_0^{\tilde{H}_{\rm SO}}, \quad (24)$$

which, like the induced transverse correlations, only depends on U. An effective longitudinal spin–orbit coupling can be defined by

$$< m,\sigma|\tilde{H}_{\rm SO}|m,\sigma> = < m,\sigma|H_{\rm SO}|m,\sigma> - U\Delta\overline{n}_{m,\sigma}. \quad (25)$$

The equation for the induced polarization is a non-linear self-consistency equation for the effective longitudinal component of the spin–orbit coupling. The non-linear relations can be written in the form of a set of coupled equations

$$\Delta\overline{n}_{m,\sigma} = -\sum_{m',\sigma'}\chi_{m,m'} < m',\sigma'|\hat{\tilde{H}}_{\rm SO}|m',\sigma'> \quad (26)$$

which formally can be solved to yield the expression for the effective longitudinal spin–orbit coupling strength

$$\tilde{\mathbf{H}}_{\rm SO} = \mathbf{H}_{\rm SO} - U\,\Delta\overline{\mathbf{n}} = [\mathbf{I} - U\chi]^{-1}\,\mathbf{H}_{\rm SO} \quad (27)$$

which is seen to be enhanced. The above results differs from those obtained by Liu *et al* [30] who do not consider off-diagonal spin correlations, so they find that only the longitudinal part of the spin–orbit coupling is enhanced. Furthermore, their enhancement is not just due to U but instead involves the combination $U' - J$ or, equivalently, $U + J$. Thus, the enhancement calculated by Liu *et al* [30] involves the Hund's rule interaction. We attribute this difference to the approximation of Liu *et al* [30] which explicitly breaks rotational-invariance and, therefore, results in only the z-component of the spin–orbit interaction being enhanced. However we do note that, since the inter-band susceptibilities are expected to be smaller than the intra-band susceptibilities, the z-component is expected to be preferentially enhanced.

The present analysis assumes the absence of inter-band coupling and so, in this treatment, the source of the enhancements of the components of the spin–orbit coupling are band specific. This leads to the speculation that the spin–orbit coupling is largest when orbital mixing and the lifting of the orbital degeneracy is smallest, i.e. at half-filling. On the other-hand when the orbital splittings are large, one may speculate that the relative size of the transverse component of the interaction ($\propto\sqrt{l(l+1)-m(m+1)}$) and the longitudinal components ($\propto m$) varies according to band-filling.

5. Discussion

Our results show that the spin–orbit interactions in strongly correlated electron materials are enhanced by the Coulomb interaction U. The spin–orbit coupling enhancement is expected to be modest compared with the enhancement of the spin susceptibility. This expectation is borne out by the spin and orbitally rotational invariant analysis, shown in the

appendix . In the absence of spin–orbit coupling, the spin susceptibility is found to be spin-rotationally invariant and is given by

$$\chi^{z,z}_{S,S} = 2\,\mu_B^2 \sum_m \frac{\left(\chi^0_{m;m} 1 - U\chi^0_{m;m}\right)}{\left[1 - J\sum_{m'}\left(\frac{\chi^0_{m';m'}}{1-U\chi^0_{m';m'}}\right)\right]}, \tag{28}$$

where $\chi^0_{m;m}$ denotes the $\omega \to 0$ and $\underline{q} \to 0$ Lindhard susceptibility for the bands with m orbital character. Likewise, the z-component of the corresponding orbital susceptibility is given by

$$\chi^{z,z}_{L,L} = 2\,\mu_B^2 \sum_{m,m'} \delta_{m,m'} \frac{m^2 \chi^0_{m,m}}{1 - (U - 2J)\chi^0_{m,m}} \tag{29}$$

and the transverse component is found to be

$$\chi^{+,-}_{L,L} = 2\,\mu_B^2 \sum_{m,m'} \delta_{m',m+1} \frac{(l(l+1) - m(m+1))\chi^{+,-}_{m,m'}}{1 - (U - 2J)\chi^{+,-}_{m,m'}}. \tag{30}$$

The above expressions are derived in the last subsection of the appendix using the rotationally invariant variational approximation. The above formulae indicate that, with the assumption that the susceptibilities are proportional to the density of states at the Fermi-energy and m-independent, the criterion for the appearance of ferromagnetic spin order reduces to

$$1 = (U + (2l+1)J)\rho(\mu), \tag{31}$$

so the Hund's rule interaction promotes ferromagnetism. On the other hand, the criterion for orbital ferromagnetism simply is given by

$$1 = (U - 2J)\rho(\mu), \tag{32}$$

which requires the presence of much stronger direct Coulomb interaction U. Although the expression in equation (31) involves the full atomic degeneracy $(2l+1)$, in a real solid with band splittings due to crystal field and spin–orbit splitting, the factor of $(2l+1)$ should be replaced by an effective orbital degeneracy [43]. Thus, the spin–orbit enhancement factor of ferromagnetic materials is expected to increase as the number of the bands, N, that participate in the Fermi-surface is reduced since use of the Stoner criterion yields an enhancement of $\sim 1 + \frac{U}{NJ}$. For ferromagnetic 5d transition metals with significant t_{2g}–e_g splittings, which sets $N \approx 2$, for which $J \sim 0.2U$ [44], one estimates that the spin–orbit enhancement factor should be approximately 2.8. Likewise, for URu_2Si_2 with $J \sim 0.33U$ [45] and $N \approx 6$, one estimates that λ is enhanced by a factor of 1.5. However, the calculations of Werwinski *et al* [22] show that, in the antiferromagnetically ordered phase, the spin–orbit splitting is about 0.8 eV which, when compared with the atomic splitting $\lambda(2l+1)/2 \sim 0.24$, indicates an enhancement of the order of 3.3. Werwinski *et al* attribute the anomalously large enhancement to near Fermi-energy nesting [46], which is known to play a significant role in the formation of the Hidden Ordered and Antiferromagnetic phases [47, 48]. The importance of spin–orbit correlations in URu_2Si_2 is indirectly indicated by RPA calculations of the paramagnetic phase which takes the tetragonal crystal structure into account, but use the atomic value of the spin–orbit splitting [28]. The RPA calculations, that did not include spin–orbit fluctuations, only produced an anisotropy in the susceptibility of 1.3 which is significantly smaller than the measured value of about 6 [29].

The concept of enhanced spin–orbit coupling might possibly also find application in the uranium monochalcogenides. The uranium monochalcogenides US, USe, and UTe have cubic crystal structures, undergo transitions to ferromagnetic phases at the temperatures of 177, 160, and 104 K, respectively [49] and exhibit giant magnetic anisotropies [50, 51]. The anisotropy is so strong that the size of the saturation magnetic moment markedly depends on the direction of the magnetization [52] and may be even responsible for introducing a slight rhombohedral distortion that occurs at the Curie temperature [53].

The concept of Coulomb correlation enhanced spin–orbit interaction has already found application in 4d–5d transition metal compounds [30] such as Sr_2RhO_4 and Sr_2IrO_4. In Sr_2RhO_4 it was found [30] that an enhanced spin–orbit coupling of about 2.15 above the bare value was necessary to account for the discrepancy between the Fermi-surface found in LDA calculations [54] with Suhubnikov–de Haas, de Haas–van Alphen [55] and ARPES [56] measurements. In Sr_2IrO_4, it has been found that the spin–orbit coupling splits the degeneracy of the d band, leading to a half-filled $j_{\rm eff} = \frac{1}{2}$ band. The effect of a small Coulomb interaction was shown to lead to a reconstruction of the normal state Fermi-surface, the formation of a gap of order 70 meV (inferred from the resistivity) which occurs across the entire Fermi-surface and the formation of a canted antiferromagnetic state in which the moments reside in the basal plane. The small magnitude of the Mott gap has been confirmed by optical absorption measurements [57], which shows that the gap decreases from 0.4 eV at $T = 10$ K to 0.08 eV at $T = 500$ K. The smallness of the gap is consistent with the claim that the material has both Mott-Hubbard and Slater character [58]. The magnetic insulating state is also rather remarkable since the canted moments are strongly locked to the oxygen octahedra, as was predicted by the LDA + SO + U calculations [17] and found in neutron diffraction experiments [59] and non-linear optical harmonic generation [60]. The calculated electronic structure shows reasonable agreement with ARPES experiments [61]. However, there are significant discrepancies in the maxima of the valence band. It has been noted that better agreement may be obtained if the spin–orbit coupling is enhanced from the value of 0.4 eV. Using the values of $U \sim 1.35$ eV and $J \sim 0.25$ appropriate for Ir [62] and assuming equal participation by the t_{2g} bands, one estimates an enhancement of 2.8 from the arguments presented here.

The realization that spin–orbit coupling strength is not simply determined by the atomic coupling, opens up the possibility that the coupling may be modified by the application of pressure, uni-axial pressure, chemical doping or magnetic fields or by applied currents, much in the same way that techniques are used to drive materials towards quantum

critical points. However, although it is feasible that a material spontaneously develops spin–orbit coupling, we consider this scenario is unlikely. The possibility of such an instability was originally uncovered by Pomeranchuk [63] in his enumeration of the instabilities of a Fermi-liquid. Recently, it was shown that an instability in the $l = 1$ channel is subjected to severe restrictions [64] but may occur in higher angular momentum channels. Likewise, the present theory predicts that the paramagnetic state should become unstable long before a transition occurs at which spin–orbit coupling is spontaneously generated. Therefore, it seems that it is much more likely that a substantial spin–orbit coupling may be created through the enhancement of the one-electron spin–orbit coupling that is guaranteed to exist by the Dirac equation.

In conclusion, we have presented a simple variational argument that indicates that the spin–orbit interaction is enhanced in strongly correlated materials and have discussed materials in which enhancements of between 2 and 3 can be found. These enhancements should become apparent in spin–orbit induced magnetic anisotropy, since the magnetic anisotropy is a high-order function of the spin–orbit coupling.

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Appendix

In the appendix we shall outline Roth's decoupling method and then indicate how the mean-field Hamiltonian can be obtained from Roth's decoupling procedure [38]. In the final subsection, we shall demonstrate how the variational method can be used to calculate various susceptibilities and show that the method maintains spin-rotationally invariance, in the absence of spin–orbit coupling.

A.1. Roth's decoupling method

The Green's function is defined as

$$G_{A_\alpha,A_\beta}(t) = -\frac{i}{\hbar} < \hat{T}\hat{A}_\alpha(t)\hat{A}^\dagger_\beta(0) > \tag{33}$$

where $\hat{T}$ is the Wick's time-ordering operator. The Martin-Schwinger hierarchy of equations of motion takes the form

$$i\hbar\frac{\partial}{\partial t}G_{A_\alpha,A_\beta}(t) = \delta(t) < \{\hat{A}_\alpha(t), \hat{A}^\dagger_\beta(0)\}_+ > + \sum_\gamma K_{\alpha,\gamma} G_{A_\gamma,A_\beta}(t) \tag{34}$$

where the interaction matrix $K_{\alpha,\gamma}$ and the fermionic operators satisfy $\hat{A}_\alpha$

$$[\hat{A}_\alpha, \hat{H}] = \sum_\gamma K_{\alpha,\gamma}\hat{A}_\gamma. \tag{35}$$

In general, the set of equations do not truncate. The truncation scheme introduced by Roth consists of evaluating the nested commutator/anticommutator

$$\{[\hat{A}_\alpha, \hat{H}], \hat{A}^\dagger_\beta\}_+ = \sum_\gamma K_{\alpha,\gamma}\{\hat{A}_\gamma, \hat{A}^\dagger_\beta\}_+ \tag{36}$$

and then taking the thermal average or expectation value in the ground state. Hence, the Fourier transformed equations of motion have the matrix form

$$\sum_\gamma \left(\hbar\omega\delta_{\alpha,\gamma} - K_{\alpha,\gamma}\right) G_{A_\gamma,A_\beta}(\omega) = < \{\hat{A}_\alpha, \hat{A}^\dagger_\beta\}_+ > . \tag{37}$$

The truncation scheme corresponds to choosing a set of operators for which the interaction matrix is Hermitean and the inhomogeneous term is positive definite. As shown by Sawada [41], this truncation procedure defines an approximate Hamiltonian $\hat{H}^{\mathrm{MF}}$ that obeys the Bogoliubov inequality

$$\begin{aligned}\Omega &= -k_B T \ \ln \ \Xi \\ &\leqslant \Omega_{\mathrm{V}} = \Omega_{\mathrm{MF}} + \mathrm{Tr}(\hat{H} - \hat{H}^{\mathrm{MF}})\hat{\rho}_{\mathrm{MF}}\end{aligned} \tag{38}$$

where $\hat{\rho}_{\mathrm{MF}}$ is the grand-canonical statistical operator derived from $\hat{H}_{\mathrm{MF}}$. Since the lower bound of Ω_{V} is Ω, Ω_{V} is a variational function.

A.2. Mean-field theory

In this section, we shall illustrate the derivation of the mean-field Hamiltonian by focussing on a particular term in the interaction Hamiltonian of equation (3). This will be followed by a presentation the various terms in the spin and orbital rotational interaction Hamiltonian. In equation (48) we shall display the mean-field interaction that is responsible for the correlations that enhance the spin–orbit interaction in equation (15).

To illustrate the derivation of the mean-field Hamiltonian shown in equation (8), which does not include spin–orbit coupling, we consider a Hamiltonian of the form

$$\begin{aligned}\hat{H} = &\sum_{i,m,i_1,m_1,\sigma} [\epsilon_m\delta_{m,m_1}\delta_{i,i_1} + t_{i,m,i_1,m_1}] f^\dagger_{i,m,\sigma} f_{i_1,m_1,\sigma} \\ &+ \frac{(U-J)}{2}\sum_{i,m\neq m_1,\sigma} f^\dagger_{i,m,\sigma} f_{i,m,\sigma} f^\dagger_{i,m_1,\sigma} f_{i,m_1,\sigma}.\end{aligned} \tag{39}$$

Since the orbital angular momentum m is measured relative to a lattice site, m is not conserved. In fact, the overlap integrals should be represented by Slater–Koster parameters [65, 66]. The commutation relation is evaluated as

$$\begin{aligned}[f_{i,m,\sigma}, \hat{H}] = &\sum_{i_1,m_1} [\epsilon_m\delta_{m,m_1}\delta_{i,i_1} + t_{i,m,i_1,m_1}] f_{i_1,m_1,\sigma} \\ &+ (U-J)\sum_{m_1\neq m} f_{i,m,\sigma} f^\dagger_{i,m_1,\sigma} f_{i,m_1,\sigma}.\end{aligned} \tag{40}$$

From this, one may restrict the Roth truncation to the set of single-particle operators

$$\hat{A}^{1}_{i,m,\sigma} = f_{i,m,\sigma}$$
$$\hat{A}^{1\dagger}_{i,m,\sigma} = f^{\dagger}_{i,m,\sigma}. \tag{41}$$

The expectation value of the double commutator is found as

$$< \{[\hat{A}^{1}_{i,m,\sigma}, \hat{H}], \hat{A}^{1\dagger}_{i',m',\sigma}\}_{+} >$$
$$= \epsilon_m \delta_{m,m'} \delta_{i,i'} + t_{i,m,i',m'} + \delta_{i,i'} \delta_{m,m'} (U-J) \sum_{m_1 \neq m} \overline{n}_{i,m_1,\sigma}$$
$$- \delta_{i,i'} (1-\delta_{m,m'})(U-J) \Phi_{i;m',\sigma;m,\sigma} \tag{42}$$

where the diagonal expectation values are denoted by

$$\overline{n}_{i,m,\sigma} = < f^{\dagger}_{i,m,\sigma} f_{i,m,\sigma} > \tag{43}$$

and the off-diagonal expectation values are denoted as

$$\Phi_{i;m,\sigma;m',\sigma} = < f^{\dagger}_{i,m,\sigma} f_{i,m',\sigma} > . \tag{44}$$

Thus, the effective Hamiltonian contains expectation values that are either diagonal or off-diagonal in the orbital index. This is generally expected to be the case since one-electron bands states can be represented by linear superpositions of atomic orbitals. A similar equation holds for the complex conjugates of the operators. The above equations are to be contrasted with the atomic Hartree–Fock equation which simply corresponds to replacing the occupation number by a site-independent average

$$f^{\dagger}_{i,m_1,\sigma} f_{i,m_1,\sigma} \rightarrow \overline{n}_{i,m_1,\sigma} \tag{45}$$

which neglects the off-diagonal terms. The quasi-particle dispersion relations $\epsilon_{\alpha,\sigma}(\underline{k})$ are then given by transforming to the Bloch representation and diagonalizing the Hermitean matrix, $\hat{H}^{\mathrm{MF}}(\underline{k})$ which has the matrix elements

$$\hat{H}^{\mathrm{MF}}_{m,m'}(\underline{k}) = \epsilon_m \delta_{m,m'} + t_{m,m'}(\underline{k}) + \delta_{m,m'} (U-J) \sum_{m_1 \neq m} \overline{n}_{m_1,\sigma}$$
$$- (U-J)(1-\delta_{m,m'}) \Phi_{m',\sigma;m,\sigma} \tag{46}$$

where the expectation values $\overline{n}_{m_1,\sigma}$ and $\Phi_{m,\sigma;m\prime\sigma}$ are to be evaluated self-consistently. The quasi-particle dispersion relations are functions of the variational parameters. An equivalent Hamiltonian $\hat{H}_{\mathrm{MF}}$ can be inferred from the dispersion relation which contains the interaction terms and their Hermitean conjugates

$$\hat{H}^{\mathrm{MF}}_{\mathrm{int}} = \sum_{i,m,i',m',\sigma} [\epsilon_m \delta_{m,m'} \delta_{i,i'} + t_{i,m,i'm'}] f^{\dagger}_{i,m,\sigma} f_{i',m',\sigma}$$
$$+ \frac{(U-J)}{2} \sum_{i,m \neq m',\sigma} \left[\hat{n}_{i,m,\sigma} \overline{n}_{i,m',\sigma} + \overline{n}_{i,m,\sigma} \hat{n}_{i,m',\sigma}\right]$$
$$- \frac{(U-J)}{2} \sum_{i,m \neq m',\sigma} \Big[f^{\dagger}_{i,m',\sigma} f_{i,m,\sigma} \Phi_{i;m,\sigma;m',\sigma}$$
$$+ \Phi_{i;m',\sigma;m,\sigma} f^{\dagger}_{i,m,\sigma} f_{i,m',\sigma} \Big] - \frac{(U-J)}{2} \sum_{i,m \neq m',\sigma} \overline{n}_{i,m,\sigma} \overline{n}_{i,m',\sigma}$$
$$+ \frac{(U-J)}{2} \sum_{i,m \neq m',\sigma} \Phi_{i;m',\sigma;m,\sigma} \Phi_{i;m,\sigma;m',\sigma}. \tag{47}$$

The mean-field interaction includes a subtraction the expectation value of the interaction between pairs of particles which avoids double-counting as expected from mean-field approximations.

Applying the above procedure to the Coulomb interaction between electrons with antiparallel spins

$$\hat{H}_{\mathrm{int}} = \frac{U}{2} \sum_{i,m,m',\sigma} \hat{n}_{i,m,\sigma} \hat{n}_{i,m',\overline{\sigma}}$$

generates the mean-field interaction

$$\hat{H}^{\mathrm{MF}}_{\mathrm{int}} = \frac{U}{2} \sum_{i,m,m',\sigma} \left[\hat{n}_{i,m,\sigma} \overline{n}_{i,m',\overline{\sigma}} + \overline{n}_{i,m,\sigma} \hat{n}_{i,m',\overline{\sigma}}\right]$$
$$- \frac{U}{2} \sum_{i,m,m',\sigma} \Big[f^{\dagger}_{i,m',\overline{\sigma}} f_{i,m,\sigma} \Phi_{i;m,\sigma;m',\overline{\sigma}}$$
$$+ f^{\dagger}_{i,m,\sigma} f_{i,m',\overline{\sigma}} \Phi_{i;m',\overline{\sigma};m,\sigma} \Big]$$
$$- \frac{U}{2} \sum_{i,m,m',\sigma} \left[\overline{n}_{i,m,\sigma} \overline{n}_{i,m',\overline{\sigma}} - |\Phi_{i;m,\sigma;m',\overline{\sigma}}|^2\right] \tag{48}$$

which includes the double-counting corrections. The Coulomb interaction U between antiparallel spins is the only interaction that generates spin and orbital flip correlations similar to the spin-flip terms in the spin–orbit coupling that enter equation (15) and are ultimately responsible for the enhancement of the spin–orbit interaction.

The mean-field interaction corresponding to the Hund's rule spin-flip interaction

$$\hat{H}_{\mathrm{int}} = -\frac{J}{2} \sum_{i,m \neq m',\sigma} f^{\dagger}_{i,m,\sigma} f_{i,m,\overline{\sigma}} f^{\dagger}_{i,m',\overline{\sigma}} f_{i,m',\sigma} \tag{49}$$

is given by

$$\hat{H}^{\mathrm{MF}}_{\mathrm{int}} = \frac{J}{2} \sum_{i,m \neq m',\sigma} \left(f^{\dagger}_{i,m,\sigma} f_{i,m',\sigma} \Phi_{i;m',\overline{\sigma},m,\overline{\sigma}} + \Phi_{i;m,\overline{\sigma},m',\overline{\sigma}} f^{\dagger}_{i,m',\sigma} f_{i,m,\sigma} \right)$$
$$- \frac{J}{2} \sum_{i,m \neq m',\sigma} \Phi_{i;m,\sigma,m',\sigma} \Phi_{i;m',\overline{\sigma},m,\overline{\sigma}} - \frac{J}{2} \sum_{i,m \neq m',\sigma}$$
$$\times \left(f^{\dagger}_{i,m,\sigma} f_{i,m,\overline{\sigma}} \Phi_{i;m',\overline{\sigma},m',\sigma} + \Phi_{i;m',\sigma,m',\overline{\sigma}} f^{\dagger}_{i,m,\overline{\sigma}} f_{i,m,\sigma} \right)$$
$$+ \frac{J}{2} \sum_{i,m \neq m',\sigma} \Phi_{i;m,\sigma,m,\overline{\sigma}} \Phi_{i;m',\overline{\sigma},m',\sigma}. \tag{50}$$

The first line describes processes which result is states that are linear superpositions of states with different orbital characters. The inter-orbital correlations enter into equation (8) since the tight-binding Hamiltonian contains orbital mixing

that stabilizes the correlations. The third line represent spin-flip interactions which contribute to the transverse spin-flip susceptibility.

The interaction

$$\hat{H}_{\rm int} = \frac{J}{2}\sum_{i,m,\sigma} f^{\dagger}_{i,m,\sigma} f_{i,m,\sigma} f^{\dagger}_{i,m,\overline{\sigma}} f_{i,m,\overline{\sigma}} \tag{51}$$

ensures the orbital rotational invariance of the exchange interaction, yields the relation $U' = U + J$. The mean-field form of the interaction is given by

$$\begin{aligned}\hat{H}^{\rm MF}_{\rm int} = {} & \frac{J}{2}\sum_{i,m,\sigma}\left(f^{\dagger}_{i,m,\sigma} f_{i,m,\sigma}\overline{n}_{i;m,\overline{\sigma}} + \overline{n}_{i;m,\sigma} f^{\dagger}_{i,m,\overline{\sigma}} f_{i,m,\overline{\sigma}}\right) \\ & - \frac{J}{2}\sum_{i,m,\sigma}\overline{n}_{i;m,\sigma}\overline{n}_{i;m,\overline{\sigma}} - \frac{J}{2}\sum_{i,m,\sigma} \\ & \times \left(f^{\dagger}_{i,m,\sigma} f_{i,m,\overline{\sigma}}\Phi_{i;m,\overline{\sigma},m,\sigma} + \Phi_{i;m,\sigma,m,\overline{\sigma}} f^{\dagger}_{i,m,\overline{\sigma}} f_{i,m,\sigma}\right) \\ & + \frac{J}{2}\sum_{i,m,\sigma}\Phi_{i;m,\sigma,m,\overline{\sigma}}\Phi_{i;m,\overline{\sigma},m,\sigma}.\end{aligned} \tag{52}$$

A.3. Spin and orbital susceptibilities

In this subsection, we shall indicate how the transverse and longitudinal susceptibilities, exhibited in equation (28) through (30), can be calculated within the variational scheme. First we shall present the spin susceptibility and show that it retains spin-rotational invariance in the absence of spin–orbit coupling. This will be followed by the derivation of the orbital susceptibility, which also exhibits rotational invariance when the system is invariant under rotations. The analysis demonstrates that the mean-field approximation is inherently spin and orbital rotationally invariant.

A.3.1. The spin susceptibility. The anomalous Zeeman interaction, $\hat{H}_{\rm Z}$, can be expressed in terms of the longitudinal and transverse parts via

$$\hat{H}_{\rm Z} = -\mu_B\left[gS^z B^z_S + \frac{g}{2}(B^+_S\hat{S}^- + B^-_S\hat{S}^+)\right]. \tag{53}$$

The total spin raising operator is given by

$$\hat{S}^+ = \sum_{i,m} f^{\dagger}_{i,m,\uparrow} f_{i,m,\downarrow} \tag{54}$$

which is diagonal in the orbital index. The lowering operator, S^-, is given by the complex conjugate expression.

The transverse susceptibility, $\chi^{+,-}_{S,S}$, can be calculated by collecting together all the single-electron spin-flip operator which are diagonal orbital angular momentum m in the mean-field Hamiltonian. These terms can be expressed as

$$-\sum_m B^-_m f^{\dagger}_{i,m,\uparrow} f_{i,m,\downarrow} \tag{55}$$

where the effective transverse field is given by

$$B^-_m = \left(\frac{g\mu_B}{2}B^-_S + U\Phi_{i;m,\downarrow;m,\uparrow} + J\sum_{m'}\Phi_{i;m',\downarrow;m',\uparrow}\right). \tag{56}$$

The first term originates from the anomalous Zeeman interaction, and the second term originates from the $m' = m$ term in the Coulomb interaction between electrons with antiparallel spins shown in equation (48). The third term comes from the mean-field terms which represent the spin-flip terms exchange interaction, displayed in equation (50), together with the correction that imposes orbital rotational invariance, shown in equation (52).

The spin-flip correlations $\Phi_{i;m',\downarrow;m,\uparrow}$ can be obtained by diagonalizing the Hamiltonian. The diagonalization can be performed by solving the coupled equations of motions for the Green's functions with the mean-field Hamiltonian. Since the mean-field Hamiltonian consists of single-electron terms, the set of equations truncate. The Fourier transform of the equations of motion become algebraic equations that depend on the tight binding elements $t_{m,m'}(\underline{k})$, $\Phi_{m,\sigma,m'\sigma}$ and $B^{\pm}_m$. For simplicity, we shall restrict our discussion to the hypothetical situation in which the orbital quantum numbers are preserved. In this case, the coupled set of equations of motion reduce to

$$\begin{aligned}(\omega - \epsilon_{m,\downarrow}(\underline{k}))G_{m,\downarrow,m,\downarrow}(\underline{k},\omega) &= 1 - B^+_m G_{m,\uparrow,m,\downarrow}(\omega) \\ (\omega - \epsilon_{m,\uparrow}(\underline{k}))G_{m,\uparrow,m,\downarrow}(\underline{k},\omega) &= -B^-_m G_{m,\downarrow,m,\downarrow}(\omega).\end{aligned} \tag{57}$$

Hence, the spin-flip Green's function is found as

$$G_{m,\uparrow,m,\downarrow}(\underline{k},\omega) = \frac{-B^-_m}{(\omega - \epsilon_{m,\downarrow}(\underline{k}))(\omega - \epsilon_{m,\uparrow}(\underline{k})) - B^+_m B^-_m} \tag{58}$$

which yields the quasiparticle energies

$$\epsilon_{m,\pm}(\underline{k}) = \frac{\epsilon_{m,\uparrow}(\underline{k}) + \epsilon_{m,\downarrow}(k)}{2} \pm \sqrt{\left(\frac{\epsilon_{m,\uparrow}(\underline{k}) - \epsilon_{m,\downarrow}(k)}{2}\right)^2 + B^+_m B^-_m}. \tag{59}$$

Therefore, the desired spin-flip correlation is given by the expectation value

$$\begin{aligned}\Phi_{m,\downarrow,m,\uparrow} &= < f^{\dagger}_{m,\downarrow} f_{m,\uparrow} > \\ &= \frac{1}{N}\sum_{\underline{k}}\left[\frac{f(\epsilon_{m,-}(\underline{k})) - f(\epsilon_{m,+}(\underline{k}))}{\sqrt{(\epsilon_{m,\uparrow}(\underline{k}) - \epsilon_{m,\downarrow}(k))^2 + 4B^+_m B^-_m}}\right]B^-_m \\ &= \chi^{-,+}_m B^-_m.\end{aligned} \tag{60}$$

In the limit $B_m \to 0$ and $T \to 0$, one finds $\chi^{+,-}_m \to \rho_m(\mu)$. More generally, the above relation takes the form

$$\Phi_{m,\downarrow,m,\uparrow} = \sum_{m'}\chi^{-,+}_{m,m'}B^-_{m'}. \tag{61}$$

On substituting the expression for the effective spin-flip field in equation (60), one finds that the spin-flip correlation is given by

$$(1 - U\chi^{-,+}_m)\Phi_{m,\downarrow,m,\uparrow} = \mu_B B^-_S\chi^{-,+}_m + J\chi^{-,+}_m\sum_{m'}\Phi_{m',\downarrow;m',\uparrow}. \tag{62}$$

The equation can be solved to yield the correlation as

$$\Phi_{m,\downarrow,m,\uparrow} = \mu_B B_S^- \frac{\chi_m^{-,+}}{1 - U\chi_m^{-,+}} \left[1 - \sum_{m'} \frac{J\chi_{m'}^{-,+}}{1 - U\chi_{m'}^{-,+}} \right]^{-1}. \quad (63)$$

Hence, one finds that the reduced spin susceptibility is

$$\sum_m \chi_m^{-,+} = \mu_B^2 \frac{\sum_m \left(\chi_m^{-,+} 1 - U\chi_m^{-,+}\right)}{\left[1 - \sum_{m'} \left(\frac{J\chi_{m'}^{-,+}}{1 - U\chi_{m'}^{-,+}}\right)\right]}, \quad (64)$$

which is identical to the result obtained from the Bethe-Salpeter equation

$$\chi_{m,m'}^{-,+} = \chi_m^{-,+} + \chi_m^{-,+} U \chi_{m,m'}^{-,+} + J\chi_m^{-,+} \sum_{m_1} \chi_{m_1,m'}^{-,+} \quad (65)$$

for the transverse dynamic spin susceptibility. Since it is customary, in linear response theory to define the induced magnetization due to the coupling with an applied field B_S^- instead of $B_S^-/2$, the transverse susceptibility includes a factor 4. Therefore, the transverse spin susceptibility is given by

$$\chi_{S,S}^{-,+} = 4\mu_B^2 \frac{\sum_m \left(\frac{\chi_m^{-,+}}{1 - U\chi_m^{-,+}}\right)}{\left[1 - \sum_{m'} \left(\frac{J\chi_{m'}^{-,+}}{1 - U\chi_{m'}^{-,+}}\right)\right]}. \quad (66)$$

The longitudinal spin susceptibility, $\chi_{S,S}^{z,z}$, can be derived by noting that the mean-field Hamiltonian [equation (8)] contains diagonal terms of the form

$$-\sum_{m,\sigma} B_{m,\sigma} f_{m,\sigma}^\dagger f_{m,\sigma}, \quad (67)$$

where the internal field $B_{m,\sigma}$ is given by

$$B_{m,\sigma} = \mu_B B_S \sigma - U \sum_{m'} \overline{n}_{m',\overline{\sigma}} - (U - J) \sum_{m' \neq m} \overline{n}_{m',\sigma} - J\overline{n}_{m,\overline{\sigma}}. \quad (68)$$

The component of the spin polarization originating from the m orbit can be expressed as

$$\sum_\sigma \sigma \overline{n}_{m,\sigma} = \sum_\sigma \sigma \int_{-\infty}^{\infty} \mathrm{d}\epsilon \rho_m(\epsilon + B_{m,\sigma}) f(\epsilon) \quad (69)$$

where the $f(\epsilon)$ that extremalizes the free-energy is the Fermi-function. The quantity χ_m is introduced via

$$\chi_m^0 = -\int_{-\infty}^{\infty} \mathrm{d}\ \epsilon \rho_m(\epsilon) \left(\frac{\partial f(\epsilon)}{\partial \epsilon}\right), \quad (70)$$

which is recognized as having the same zero field value as the quantity $\chi_m^{+,-}$ introduced previously. On expanding the right-hand side of the spin polarization in terms of the effective field, one finds the equation

$$\sum_\sigma \sigma \overline{n}_{m,\sigma} (1 - U\chi_m^0) = 2\,\mu_B\, B_S \chi_m^0 + J\chi_m^0 \sum_{m',\sigma} \sigma \overline{n}_{m',\sigma}. \quad (71)$$

This equation can be solved to yield the spin polarization

$$\sum_\sigma \sigma \overline{n}_{m,\sigma} = 2\,\mu_B\, B_S \frac{\left(\chi_m^0 1 - U\chi_m^0\right)}{\left[1 - \sum_{m'} \left(\frac{J\chi_{m'}^0}{1 - U\chi_{m'}^0}\right)\right]}. \quad (72)$$

Hence, one identifies the longitudinal spin susceptibility as

$$\chi_{S,S}^{z,z} = 2\mu_B^2 \frac{\sum_m \left(\frac{\chi_m^0}{1 - U\chi_m^0}\right)}{\left[1 - \sum_{m'} \left(\frac{J\chi_{m'}^0}{1 - U\chi_{m'}^0}\right)\right]}. \quad (73)$$

Comparison of the longitudinal with the transverse spin susceptibility confirms that $\chi_{S,S}^{+,-} = 2\chi_{S<S}^{z,z}$ which indicates that spin-rotational invariance is preserved by the mean-field approximation.

A.3.2. The orbital susceptibility. The Zeeman interaction with the orbital moment can be expressed as

$$\hat{H}_Z = -\mu_B \left[B_L^z \hat{L}^z + \frac{1}{2} \left(B_L^+ \hat{L}^- + B_L^- \hat{L}^+ \right) \right]. \quad (74)$$

By contrast to the spin susceptibilities, the orbital susceptibilities are anisotropic in the absence of spin–orbit coupling.

The transverse orbital susceptibility can be calculated by noting that the transverse field which acts on the orbital B_L can be combined with the induced orbital polarizations to yield an effective transverse interaction. The effective transverse orbital interaction conserves the spin quantum number and is written as

$$\hat{H} = -\sum_{i,m,\sigma} [f_{i,m+1,\sigma}^\dagger f_{i,m,\sigma} B_m^- + f_{i,m,\sigma}^\dagger f_{i,m+1,\sigma} B_m^+] \quad (75)$$

where the effective transverse orbital field is given by

$$B_m^- = \sqrt{l(l+1) - m(m+1)} B_L^- + (U - J)\Phi_{i;m,\sigma,m+1,\sigma} - J\Phi_{i;m,\overline{\sigma},m+1,\overline{\sigma}}. \quad (76)$$

Since the differential transverse orbital susceptibility is calculated in the zero field limit, one may assume spin-rotational invariance. Thus, the effective interaction is spin-independent and is given by

$$B_m^- = \sqrt{l(l+1) - m(m+1)} B_L^- + (U - 2J)\Phi_{i;m,m+1} \quad (77)$$

where we have dropped the spin indices. The equations of motion for the Green's functions can be written as a matrix equation

$$\left(\hbar\omega \hat{I} - \hat{H}^{\mathrm{MF}}(\underline{k})\right) \underline{G}(\omega, \underline{k}) = \underline{\delta} \quad (78)$$

where $\underline{\delta}$ is a unit vector. If the hopping Hamiltonian is diagonal in m, the matrix is a Hermitean tridiagonal matrix. The determinant is denoted by $D_{\underline{k}}(\omega)$

$$D_{\underline{k}}(\omega) = |\hbar\omega \hat{I} - \hat{H}^{\mathrm{MF}}(\underline{k})|. \quad (79)$$

The quasiparticle dispersion relations are found from the secular equation

$$D_{\underline{k}}(\omega) = 0. \quad (80)$$

Since the cofactor of the mth diagonal element is given by

$$-\frac{\partial}{\partial \epsilon_m} D_{\underline{k}}(\omega) \tag{81}$$

where ϵ_m is the binding energy of the mth atomic orbit, the diagonal Green's function $G_{m,m}(\omega, \underline{k})$ is found as

$$G_{m,m}(\omega, \underline{k}) = -\frac{\partial}{\partial \epsilon_m} \ln D_{\underline{k}}(\omega). \tag{82}$$

The diagonal Green's function allows the direct calculation of the thermal average $\overline{n}_m$. The off-diagonal Green's function is given by the derivative

$$G_{m+1,m}(\omega, \underline{k}) = \frac{\partial}{\partial B_m^+} \ln D_{\underline{k}}(\omega). \tag{83}$$

Furthermore, due to the tridiagonal nature of the determinant, it only depends linearly on the combination $B_m^+ B_m^-$, which leads to the expectation value of the off-diagonal Green's function being given by

$$\Phi_{m,m+1} = \chi_m^{-,+} B_m^-. \tag{84}$$

Hence, on substituting the expression for the effective field B_m^-, one obtains the equation

$$\Phi_{m,m+1} = \chi_m^{-,+} \left[\sqrt{l(l+1) - m(m+1)} B_L^- + (U - 2J)\Phi_{i;m,m+1} \right] \tag{85}$$

which results in the transverse orbital susceptibility being given by

$$\chi_{L,L}^{-,+} = 2\mu_B^2 \sum_{m,m'} \delta_{m',m+1} \frac{(l(l+1) - m(m+1))\chi_{m,m'}^{-,+}}{1 - (U - 2J)\chi_{m,m'}^{-,+}}, \tag{86}$$

where the factor of two comes from the spin degeneracy.

The longitudinal orbital susceptibility, $\chi_{L,L}^{z,z}$, can be found by identifying a single-electron interaction with the z-component of the orbital magnetic field B_L

$$\hat{H}_L = -\sum_{m,\sigma} f_{m,\sigma}^\dagger f_{m,\sigma} \left[\mu_B B_L m - U \sum_{m'} \overline{n}_{m',\overline{\sigma}} - (U - J) \sum_{m' \neq m} \overline{n}_{m',\sigma} - J \overline{n}_{m,\overline{\sigma}} \right]. \tag{87}$$

In the absence of B_S^z, the system is spin-rotationally invariant. Therefore, the expectation values are spin independent

$$\overline{n}_{m,\sigma} = \overline{n}_{m,\overline{\sigma}} = \overline{n}_m. \tag{88}$$

Under this condition, the effective interaction with the orbital field is given by

$$\hat{H}_L = -\sum_{m,\sigma} f_{m,\sigma}^\dagger f_{m,\sigma} B_m \tag{89}$$

where we have introduced an effective orbital field

$$B_m = \left[\mu_B B_L m + (U - 2J)\overline{n}_m - (U - \frac{J}{2}) N_{\mathrm{T}} \right] \tag{90}$$

and where N_{T} is the total number of electrons. Hence, the last term in B_m is independent of the applied field B_L. For a system that is invariant under rotations, the average value of the occupation of the mth orbital is given by

$$\overline{n}_m = \int_{-\infty}^{\infty} \mathrm{d}\epsilon \rho_m(\epsilon + B_m) f(\epsilon) \tag{91}$$

where $\rho_m(\epsilon)$ is the single-particle density of states for the mth orbital. On expanding the right-hand side in terms of the field dependent parts of the thermally average occupation numbers, one finds

$$\Delta \overline{n}_m = \chi_m \left[\mu_B B_L + (U - 2J)\Delta \overline{n}_m \right] \tag{92}$$

in which we have introduced the susceptibility

$$\chi_m^0 = -\int_{-\infty}^{\infty} \mathrm{d}\epsilon \rho_m(\epsilon) \left(\frac{\partial f(\epsilon)}{\partial \epsilon} \right). \tag{93}$$

The above equations are solved for the orbital polarization

$$\Delta \overline{n}_m = \mu_B B_L \frac{\chi_m^0}{1 - (U - 2J)\chi_m^0}. \tag{94}$$

If the z-component of the orbital magnetization is defined by

$$M_L^z = 2\mu_B \sum m \Delta \overline{n}_m \tag{95}$$

the longitudinal orbital susceptibility is given by

$$\chi_{L,L}^{z,z} = 2\mu_B^2 \sum_m \frac{m^2 \chi_m^0}{1 - (U - 2J)\chi_m^0}. \tag{96}$$

Hence, the orbital polarization is enhanced susceptibility is enhanced by the Coulomb and the exchange interaction in the combination $(U - 2J)$.

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