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Enhanced Spin Orbit Interaction in Actinides

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Abstract. We investigate the effect of many-body interactions on the spin-orbit coupling of anisotropic metals. We use the Underscreened Anderson Lattice Model that was proposed to describe actinide compounds. The Coulomb interactions induce off-diagonal correlations that enhance the components of the spin-orbit coupling. Modest values of the Coulomb interaction U can significantly enhance the spin-orbit coupling and effect the electronic spectrum. The enhancements are most pronounced for systems that are on the verge of magnetic instabilities. The enhancement is anisotropic in crystals with non-cubic symmetries and can lead to giant magnetic anisotropies in paramagnetic states.

1. Introduction

Actinide materials exhibit strong electronic correlations and spin-orbit interactions. The 5f wave functions are spatially extended and often exhibit giant magnetic anisotropies, such as in the cubic ferromagnetic uranium monochalcogenides [1], tetragonal URu_2Si_2 [2, 3] and ordered phase of α-plutonium [4, 5]. The giant magnetic anisotropies persist in non-magnetic states. It has been proposed that the anisotropies are due to the enhanced spin-orbit coupling strengths caused by electron correlations [6]. The Underscreened Kondo Model was formulated to describe the properties of actinide materials, such as the uranium monochalcogenides where the resistivity shows logarithmic temperature variations over several decades of temperature [7] but order ferromagnetically at temperatures of the order of 100 K, and have ordered magnetic moments of the order of a Bohr magneton [1]. This has been described in terms of the underscreened Kondo Model [8, 9] in which the Kondo effect only partially screens the local f moment, and the residual local moments order at low temperatures. In the simplest version of the underscreened Anderson Lattice Model, the atomic f orbitals are degenerate with the result that only a single combination of the f orbitals hybridizes and participates in the Kondo screening. The remaining f bands do not hybridize and host the formation of the residual low temperature local moments. Here we shall consider the effect of spin-orbit interaction in the underscreened Anderson Lattice Model, and find that the itinerant character of the model and the Coulomb interaction leads to an enhanced spin-orbit coupling which is anisotropic in non-cubic materials [10].

2. The Underscreened Anderson Lattice Model

We consider a simple orthorhombic crystal, in which a set of A atoms are located at the $(0, 0, 0)$ sites and the second set of B atoms are located at $(\frac{a_x}{2}, \frac{a_y}{2}, \frac{a_z}{2})$ sites. The A atoms host the itinerant conduction states, whereas the B sites host f states that are subjected to spin-orbit

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coupling and strong electron-electron interactions. We consider the conduction band as non-degenerate and the f orbitals as triply-degenerate. The Hamiltonian can be represented as the sum of three terms,

$$\hat{H} \;=\; \hat{H}_A \;+\; \hat{H}_B \;+\hat{H}_{AB} \tag{1}$$

where $\hat{H}_A$ describes the Hamiltonian for the conduction band and is given by

$$\hat{H}_A \;=\; \sum_{\underline{k},\sigma} \epsilon_a(\underline{k})\; c^\dagger_{\underline{k},\sigma}\; c_{\underline{k},\sigma} \tag{2}$$

in which $c^\dagger_{\underline{k},\sigma}$ and $c_{\underline{k},\sigma}$, respectively create and annihilate an electron with spin σ in the conduction band with Bloch wave vector $\underline{k}$. The Hamiltonian for the electrons in the orbitals of the B atoms are described by the sum of a tight-binding Hamiltonian $\hat{H}_0$, the spin-orbit interaction, $\hat{H}_{SO}$ and $\hat{H}_{int}$ which describes the Coulomb interactions

$$\hat{H}_B \;=\; \hat{H}_0 \;+\; \hat{H}_{SO} \;+\; \hat{H}_{int} \tag{3}$$

The tight-binding Hamiltonian for the localized orbitals, $\hat{H}_0$, is given by

$$\hat{H}_0 \;=\; \sum_{\underline{k},\beta,\sigma} \epsilon_\beta(\underline{k})\; f^\dagger_{\underline{k},\beta,\sigma}\; f_{\underline{k},\beta,\sigma} \tag{4}$$

where $\beta = x, y, z$, and where $f^\dagger_{\underline{k},\beta,\sigma}$ and $f_{\underline{k},\beta,\sigma}$, respectively create or annihilate an electron of spin σ, $\sigma = \pm 1$, and Bloch wave-vector $\underline{k}$ in the β-th orbital on the B atom. In the cubic harmonic Bloch representation, the spin-orbit interaction has the form

$$\begin{aligned}
\hat{H}_{SO} \;=\; & \frac{\lambda_z}{2} \sum_{\underline{k},\sigma} i\;\sigma \left[\; f^\dagger_{\underline{k},y,\sigma} f_{\underline{k},x,\sigma} \;-\; f^\dagger_{\underline{k},x,\sigma} f_{\underline{k},y,\sigma} \;\right] \\
& - \frac{\lambda_y}{2} \sum_{\underline{k},\sigma} \sigma \left[\; f^\dagger_{\underline{k},z,\sigma} f_{\underline{k},x,\overline{\sigma}} \;+\; f^\dagger_{\underline{k},x,\overline{\sigma}} f_{\underline{k},z,\sigma} \;\right] \\
& + \frac{\lambda_x}{2} \sum_{\underline{k},\sigma} i \left[\; f^\dagger_{\underline{k},z,\sigma} f_{\underline{k},y,\overline{\sigma}} \;-\; f^\dagger_{\underline{k},y,\overline{\sigma}} f_{\underline{k},z,\sigma} \;\right]
\end{aligned} \tag{5}$$

where we have introduced different coupling strengths in anticipation of the result that the Coulomb enhancement will break the atomic symmetry. The interaction between the f electrons is denoted as $\hat{H}_{int}$. The interaction is composed of the on-site Coulomb interaction U and the Hund's rule interaction J between the electrons in an atomic shell. In principle, the atomic interactions U and J should depend on the pair of orbital quantum numbers m and m'. We shall neglect this dependence but instead require that the interaction be invariant under both spin and orbital rotations [11]. In the tetragonal basis, the interaction has the form

$$\begin{aligned}
\hat{H}_{int} \;=\; & \frac{1}{2} \sum_{i,\alpha,\beta,\sigma} U\; \hat{n}_{i,\alpha,\sigma}\; \hat{n}_{i,\beta,\overline{\sigma}} \\
& + \frac{1}{2} \sum_{i,\alpha\neq\beta,\sigma} (U - J)\; \hat{n}_{i,\alpha,\sigma}\; \hat{n}_{i,\beta,\sigma} \\
& - \frac{1}{2} \sum_{i,\alpha\neq\beta,\sigma} J\; f^\dagger_{i,\alpha,\sigma}\; f_{i,\alpha,\overline{\sigma}}\; f^\dagger_{i,\beta,\overline{\sigma}}\; f_{i,\beta,\sigma} \\
& + \frac{1}{2} \sum_{i,\alpha,\sigma} J\; \hat{n}_{i,\alpha,\sigma}\; \hat{n}_{i,\alpha,\overline{\sigma}}
\end{aligned} \tag{6}$$

where the sums over α and β run through x, y, z. Thus, the strength of the intra-orbital Coulomb interaction, (i.e. $\alpha = \beta$) is given by $U' = U + J$ and, therefore, is distinct from the inter-orbital interaction U. The hybridization term that couples the A and B orbitals on nearest-neighbor atoms has the form

$$\hat{H}_{AB} \;=\; \sum_{\underline{k},\beta,\sigma} \; i \;\; V_\beta(\underline{k}) \; \Big[\; c^\dagger_{\underline{k},\sigma} f_{\underline{k},\beta,\sigma} \;-\; f^\dagger_{\underline{k},\beta,\sigma} c_{\underline{k},\sigma} \; \Big] \tag{7}$$

where the k-dependent hybridization matrix elements are given by

$$V_\beta(\underline{k}) \;=\; 8 \; V_{AB} \; \frac{a_\beta}{\sqrt{\sum_\gamma a_\gamma^2}} \; \sin \frac{k_\beta a_\beta}{2} \prod_{\alpha \neq \beta} \Big(\cos \frac{k_\alpha a_\alpha}{2} \Big) \tag{8}$$

in which V_{AB} is the Slater-Koster parameter $t^\sigma_{A,B}(R)$ where R is the distance between the atoms. The hybridization is primarily responsible for the width of the 5f band.

3. Results and Discussion

The electrons in the hybridized bands are subjected to Coulomb and spin-orbit scattering which interfere constructively. In the mean-field approximation, the induced Coulomb correlations have the same symmetries as the components of the spin-orbit interaction and produce an anisotropic enhancement

$$\begin{aligned} -\, i\, \sigma \, \frac{\lambda_z}{2} \;&=\; -\, i\, \sigma \, \frac{\lambda_0}{2} \;-\; U \;<\! f^\dagger_{i,y,\sigma} f_{i,x,\sigma} \!> \\ -\, \sigma \, \frac{\lambda_y}{2} \;&=\; -\, \sigma \, \frac{\lambda_0}{2} \;-\; U \;<\! f^\dagger_{i,z,\sigma} f_{i,x,\overline{\sigma}} \!> \\ -\, i \, \frac{\lambda_x}{2} \;&=\; -\, i \, \frac{\lambda_0}{2} \;-\; U \;<\; f^\dagger_{i,z,\sigma} f_{i,y,\overline{\sigma}} \!> \end{aligned} \tag{9}$$

The off-diagonal spin-flip correlations are obtained from the respective Green's functions

$$\begin{aligned} G_{z,\sigma,x,\overline{\sigma}}(\omega, \underline{k}) \;&=\; -\, \sigma \, \Big[\, \frac{\lambda_y}{2} \, \Big((\omega - \epsilon_y)(\omega - \epsilon_a) - V_y^2 \Big) - \frac{\lambda_x \lambda_z}{4} \; (\omega \; - \epsilon_a) \, \Big] / D_4(\omega, k) \\ &\quad + \, i \, \sigma \, \frac{\lambda_x}{2} \, \frac{V_x \; V_y}{D_4(\omega, \underline{k})} \end{aligned} \tag{10}$$

and

$$\begin{aligned} G_{y,\overline{\sigma},z,\sigma}(\omega, \underline{k}) \;&=\; -\, i \, \Big[\, \frac{\lambda_x}{2} \, \Big((\omega - \epsilon_x)(\omega - \epsilon_a) - V_x^2 \Big) - \frac{\lambda_y \lambda_z}{4} \; (\omega \; - \epsilon_a) \, \Big] / D_4(\omega, k) \\ &\quad + \, \sigma \, \frac{\lambda_y}{2} \, \frac{V_x \; V_y}{D_4(\omega, \underline{k})} \end{aligned} \tag{11}$$

where $D_4(\omega, \underline{k})$ is the secular determinant which determines the degenerate quasiparticle dispersion relations expected for a time-reversal invariant centro-symmetric material. The enhancement of the α-component of the spin-orbit interaction contains terms which are proportional $\lambda_\alpha V_x V_y$ that are expected to vanish for a centro-symmetric system. Similar terms also appear in the expression for the directionally dependent magnetic susceptibility which, if non-zero, would produce an anisotropy along the (1,1,1) direction for a cubic system. We note that, for compounds with strong hybridization and spin-orbit coupling, the formation of a magnetic moment can induce a slight structural distortion that stabilizes the $(1, 1, 1)$ direction as an easy-axis. This is in accord with recent observations on ferromagnetic US [12].

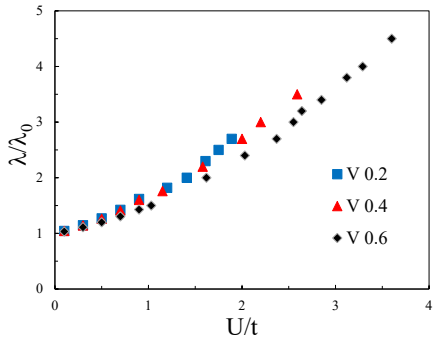

Figure 1. The ratio of the enhanced spin orbit coupling to the atomic value, as a function of U/t for various values of the hybridization V/t.

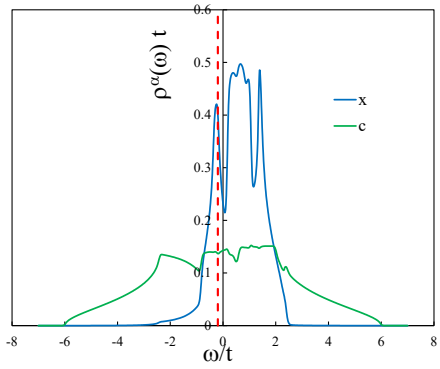

Figure 2. The partial conduction band and f sub-band density of states for $V/t = 0.15$ and $U/t \sim 1$. The Fermi-energy is indicated by the dashed vertical line.

For cubic compounds, the spin-orbit interaction is isotropic. The enhancement of the spin orbit coupling λ/λ_0 in the paramagnetic state is shown as a function of U for various values of the hybridization in fig(1). The conduction band width has been set at $12t$, the f tight-binding parameters have been set at 0.3 t, the atomic spin-orbit coupling is $\lambda_0 = 0.24t$ and $J = U/6$. It is seen that the enhancement increases with U and is maximum at the critical value above which the system becomes magnetic. The critical value of U increases when V is increased. The largest enhancements are found at the quantum critical point, and the are maximal for large V. The single particle f and conduction and f band partial densities of states are shown in fig(2) for $U/t \approx 1$. It is seen that the enhanced spin-orbit coupling produces pseudo-gaps in the f density of states at energies associated with avoided band crossings. The enhancement of the spin-obit coupling is associated with the increased stability that occurs when the chemical potential is inside a pseudo-gap. Hence, the enhanced spin-orbit coupling is expected to be manifest in the density of states as probed in ARPES/BIS experiments.

4. References

[1] Lander GH, Brooks MSS, Lebech B, Brown PJ, Vogt O and Mattenberger K 1990 Appl. Phys. Lett. **57**, 989-91.
[2] Palstra TTM, Menovsky AA, van den Berg J, Dirkmaat AJ, Kes PH, Nieuwenhuys GJ and Mydosh JA 1985 Phys. Rev. Lett. **55**, 2727-30.
[3] Werwinski M, Rusz J, Mydosh JA and Oppeneer PM 2014 Phys. Rev. B, **90**, 064430 (2014).
[4] Kutepov AL and Kutepova SG, 2003 J. Phys. Cond. Mat. **15**, 2607-24.
[5] Butterfield MT, Tobin JG, Teslich NE, Bliss RA, Wall MA, McMahan AK, Chung BW, Schwartz AJ and Kutepov AL 1996 Matl. Res. Soc. Symp. Proc. **893**, 95.
[6] Riseborough PS, Magalhaes SG, Calegari EJ and Cao G 2020 J. Phys., Cond. Mat. **32**, 4455601.
[7] Schoenes J, Frick B and Vogt O 1984 Phys. Rev. B **30**, 6578-85.
[8] Perkins NB, Nunez-Regueiro MD, Coqblin B and Iglesias JR 2007 Phys. Rev. B, **76**, 125101.
[9] Thomas C, Simoes ASD, Lacroix C, Iglesias JR and Coqblin B, 2014 J. Mag. Mag. Mat. **372**, 247-52.
[10] Yuan X, Riseborough PS, Calegari EJ and Magalhaes SG 2021 Electron. Struct. **3**, 024009.
[11] Dworin L and Narath A 1970 Phys. Rev. Lett. **25**, 1287-91.
[12] Poudel N, Jeffries J, Gofryk K 2021 Phys. Rev. B, **104** 014417.