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Elastic anomaly of heavy fermion systems in a crystalline field

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Abstract. An elastic anomaly, observed in the heavy Fermi liquid state of Ce alloys (for example, $CeCu_6$ and CeTe), is analysed by using the infinite-U Anderson lattice model. Four atomic energy levels are assumed for f-electrons. Two of them are mutually degenerate. A small crystalline splitting 2Δ is assumed between two energy levels. The fourfold degenerate conduction bands are also considered in the model. We solve the model using the mean-field approximation to slave bosons, changing the Fermi energy in order to keep the total electron number constant. The non-zero value of the mean field of the slave bosons persists over temperatures much higher than the Kondo temperature. This is an effect of the constant electron number. Next, the linear susceptibility with respect to Δ is calculated in order to obtain the renormalized elastic constant. The resulting temperature dependence of the constant shows a downward dip. We discuss the relation of our findings to experimental data.

1. Introduction

In general, the atomic f-level of heavy fermion Ce compounds is split by a crystalline field into multi-sublevels [1]. The original f-level of the degeneracy, 14, is first split into $j = 5/2$ and $7/2$ levels by the l–s interaction. The $j = 7/2$ level is about 10^3 K higher than the $j = 5/2$ level, so the main contribution comes from the $j = 5/2$ level, and the $j = 7/2$ level can be neglected. Second, the $j = 5/2$ level splits into several multiplets in the presence of the crystalline field. For example, in the cubic field, the $j = 5/2$ level is composed of two sublevels of degeneracy two and four, and in the tetragonal field it splits into three Kramers doublets. When the ground level is doubly degenerate, the lowest f-level is nearly half filled because the number of the 4f-electron is close to 0.9. The splitting width is normally 10^{1-2} K, so the temperature dependence of the physical quantities reflects the effect of the thermal excitations among the sublevels.

The effect of the crystalline field on the single-site Kondo system has been a recent topic in the theory of dilute magnetic alloys. The Anderson model or the Coqblin–Schrieffer model, with the inclusion of the crystalline field and the anisotropy of the mixing interaction, is solved by the use of the Bethe ansatz technique [2] and the self-consistent renormalization [3]. The result shows the variation of the Kondo temperature T_K with temperature. It comes from the change of the impurity scattering channel. For example, we imagine the case where there are three Kramers doublets with splitting Δ_1 and Δ_2. In the temperature range $T < \Delta_1$, the three levels act independently in the scattering but, in the range $\Delta_1 < T < \Delta_2$, the lower two levels behave as if they are one level of degeneracy four, because the thermal fluctuation has a larger width than Δ_1. In the same way, in the range $T > \Delta_2$, all three levels look like a sixfold degenerate level. It has been postulated that the above features are exhibited in the temperature dependence of the magnetic susceptibility.

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Experimentally, the heavy fermion systems show elastic anomalies related to valence instabilities at low temperatures. The most striking effect is the softening of the elastic constants below the Kondo temperature, which are observed prominently in $CeAl_3$ [4], $CeCu_6$ [5] and $CeRu_2Si_2$ [6]. The temperature dependences of the elastic constants have been theoretically fitted quantitatively [7] by using the electron–phonon coupling derived from the ansatz that the mixing strength depends on the volume of the crystal. The same ansatz was highly successful in the theoretical description [8,9] of the Kondo volume collapse transition between α-Ce and γ-Ce. These elastic anomalies are related to the overall instability of the valence mainly due to the change of the mixing interaction. They are observed in the wide temperature range $0 < T < T_K$ and are rather insensitive to the detailed structures of the electronic band structures.

However, when we look at detailed temperature dependences of the elastic constants, several compounds show anomalies which have been interpreted as the crystalline field effect. For example, the elastic constant C_{33} of $CeCu_6$ [10] has a dip at about 10 K. This might be due to a splitting larger than the Kondo temperature 4 K. The constant $(C_{11}-C_{12})/2$ of CeTe [11] shows an apparent dip at about 15 K. The ground-state levels of Ce ions split into the Γ_7 Kramers doublet and the Γ_8 quartet states. The Γ_7 states are the ground states. There is a splitting of 30 K between Γ_7 and Γ_8 states. This is the origin of the dip.

The main purpose of this paper is to present a microscopic calculation in order to look at how the degeneracy structure and crystalline field appear in the elastic properties. The formalism is independent of the real band structures and degeneracy structures. Therefore, the results should be general enough for heavy fermion systems. We simply assume four quantum numbers for f-electrons. Two of them have the same atomic energy level; thus two different atomic levels are assumed. The difference between them, smaller than T_K, is the width of the crystalline field splitting. Four conduction bands are assumed. The total electronic system is described by the infinite-U Anderson lattice Hamiltonian by the slave boson method. The model is solved by the mean-field approximation. The formalism is explained in section 2.

Firstly, we show mean-field solutions and see that the mean-field value of slave bosons does not vanish even at $T > T_K$. This interesting property has been discussed previously [12, 13]. It is due to the fact that the Fermi level is varied in order to keep the total electron number constant. This is shown in section 3.

Secondly, we calculate the linear susceptibility with respect to the crystalline field splitting. At lower temperatures, the susceptibility has a structure related to the crystalline field effect. The high-temperature susceptibility does not depend on the splitting width, due to the large thermal excitation. There is an upward dip in the temperature dependence. The dip originates from the high degeneracy, and its position moves with the splitting width.

Next, we assume an empirical relation between an elastic constant and the susceptibility. The form of the relation is assumed by taking account of the quadrupolar response theory [14]. As the coupling constant between the lattice and the crystalline field is unknown, we should treat it as a type of parameter. We show the temperature dependences of elastic constants for several choices of the coupling. We will discuss relevant parameters for the elastic anomaly, i.e. the downward dip that is observed in the constant $(C_{11} - C_{12})/2$ of CeTe [11]. The susceptibility and elastic constant are reported and discussed in section 4.

We conclude with several remarks in section 5.

2. Formalism

We consider the infinite-U Anderson lattice model in the slave boson method. The model has the following form:

$$H = \sum_i \left((E_f - \Delta) \sum_{l=1,2} f_{i,l}^\dagger f_{i,l} + (E_f + \Delta) \sum_{l=3,4} f_{i,l}^\dagger f_{i,l} \right) + \sum_{k,l=1-4} \varepsilon_k c_{k,l}^\dagger c_{k,l} + V \sum_{i,l=1-4} (f_{i,l}^\dagger c_{i,l} b_i + b_i^\dagger c_{i,l}^\dagger f_{i,l}) + \sum_i \lambda_i \left(\sum_{l=1-4} f_{i,l}^\dagger f_{i,l} + b_i^\dagger b_i - 1 \right) \quad (1)$$

where $f_{i,l}$ is an annihilation operator of the f-electron of the lth orbital at the ith site, $c_{k,l}$ is an operator of the conduction electron with wave number k, and b_i is an operator of the slave boson which indicates the unoccupied state at the f-orbital. The atomic energy of the first and second orbitals of f-electrons is $E_f - \Delta$, and that of the third and fourth orbitals is $E_f + \Delta$. A crystalline field splitting 2Δ is assumed between atomic energies of the two groups of f-electrons. For the conduction electrons, the same quantum number is assumed as that of the f-electrons. We use the square density of states, $\rho \equiv 1/ND$, which extends over the energy region, $-D < \varepsilon_k < (N-1)D$, where $N = 4$ is the total number of quantum states. This assumes that the combination $N\rho V^2$, which appears in the $1/N$ expansion, is independent of N. Therefore, the mean-field theory becomes exact as $N \to \infty$. The third term in the Hamiltonian is the mixing interaction between f- and c-electrons, V being the interaction strength. The last term limits the maximum number of f-electrons per site up to unity. This could be realized by the constraint

$$\sum_{l=1-4} f_{i,l}^\dagger f_{i,l} + b_i^\dagger b_i = 1$$

with the Lagrange multiplier field λ_i.

This model is treated within the mean-field approximation: $\langle b_i \rangle = r$, $\langle b_i^\dagger b_i \rangle = r^2$ and $\lambda_i = \lambda$ (a site-independent real value). These mean-field parameters are determined by solving the following coupled equations. First, the constraint condition

$$\frac{1}{2D} \int dE \frac{\tilde{V}^2}{(\tilde{E}_f - \Delta - E)^2} f(E-\mu) + \frac{1}{2D} \int dE \frac{\tilde{V}^2}{(\tilde{E}_f + \Delta - E)^2} f(E-\mu) + r^2 = 1. \quad (2)$$

Second, the self-consistency condition for r:

$$\frac{1}{2D} \int dE \frac{V^2}{E - \tilde{E}_f + \Delta} f(E-\mu) + \frac{1}{2D} \int dE \frac{V^2}{E - \tilde{E}_f - \Delta} f(E-\mu) + \lambda = 0. \quad (3)$$

And third, the conservation condition of electron number n_{el}:

$$\frac{1}{2D} \int dE \left(1 + \frac{\tilde{V}^2}{(\tilde{E}_f - \Delta - E)^2} \right) f(E-\mu) + \frac{1}{2D} \int dE \left(1 + \frac{\tilde{V}^2}{(\tilde{E}_f + \Delta - E)^2} \right) f(E-\mu) = n_{el} \quad (4)$$

where $f(x) = 1/[\exp(x/T) + 1]$ is the Fermi distribution function, $\tilde{E}_f = E_f + \lambda$ is the effective f-level, and $\tilde{V} = rV$ is the effective mixing interaction. The integrations are performed over the entire energy region of the bands. The three equations are solved numerically for the variables, r, λ and the Fermi level μ. In addition, the values at $T = 0$ can be obtained analytically.

3. Solution

Equations (2)–(4) are solved numerically for the parameters $D = 5 \times 10^4$ K, $V = 7500$ K, $E_f = -10^4$ K, and $n_{el} = 1.9$ as the typical values. The splitting parameter Δ is increased from zero, but the magnitude is taken below the Kondo temperature $T_K = \tilde{E}_f - \mu$.

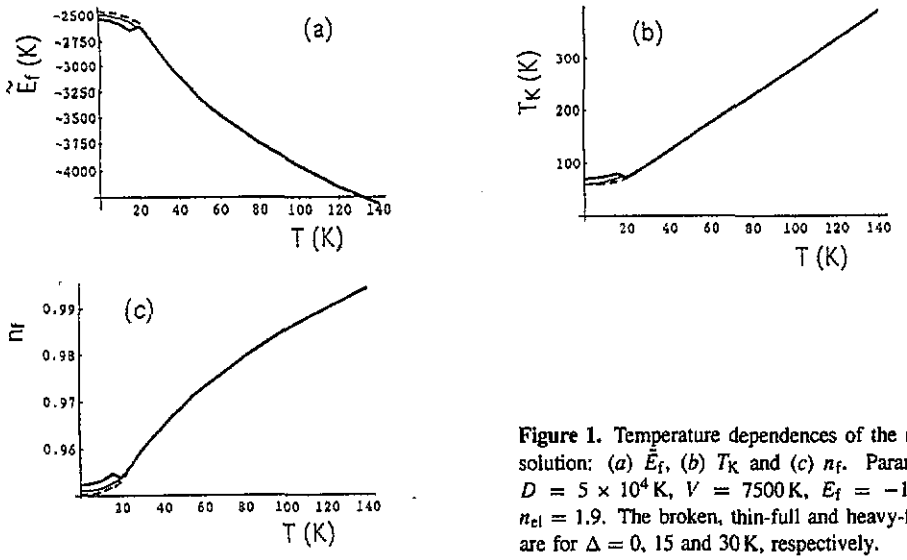

Figure 1. Temperature dependences of the mean-field solution: (*a*) $\tilde{E}_f$, (*b*) T_K and (*c*) n_f. Parameters are $D = 5 \times 10^4$ K, $V = 7500$ K, $E_f = -10^4$ K, and $n_{el} = 1.9$. The broken, thin-full and heavy-full curves are for $\Delta = 0$, 15 and 30 K, respectively.

Figure 1 shows the temperature dependences of parameters. Figures 1 (*a*), (*b*) and (*c*) show the variations of $\tilde{E}_f$, T_K and the number of f-electrons per site n_f, respectively. The data for $\Delta = 0$, 15 and 30 K are shown by the broken, thin-full and thick-full curves, respectively. This convention applies to all the figures in this paper. As the temperature increases, the order parameter r decreases, so that $n_f = 1 - r^2$ increases. The quantity r does not vanish even though the temperature is much higher than T_K at $T = 0$. This is the effect of the change of the Fermi level μ to keep the total electron number constant. This effect has been reported previously [12, 13]. As there is an increase of n_f, $\tilde{E}_f$ decreases, which means a reduced itinerancy of f-electrons. At high temperatures, $T \gg \Delta$, the three curves becomes almost identical, owing to the large temperature excitation across the split 2Δ. At low temperatures, the excitation energy is limited by the smaller distance from the Fermi level to the gap of the bands $l = 1, 2$. This results in the increased value of n_f when the crystalline field is switched on.

We can derive the analytical expressions of parameters at $T = 0$. A small crystalline field splitting is assumed for the derivation. The results are

$$T_K(\Delta) \equiv \tilde{E}_f - \mu = (T_K^2(0) + \Delta^2)^{1/2} \tag{5}$$

$$r^2 \simeq DT_K^2(0)/V^2 T_K(\Delta) \tag{6}$$

where $T_K(0) = D\exp[-D(\mu - E_f)/V^2]$ is the Kondo temperature for $\Delta = 0$. These expressions accord with the numerical results that both T_K and n_f increase as Δ increases.

4. Elastic anomaly in low temperatures

We shall now discuss the change of the elastic property of heavy fermions due to the crystalline field splitting at temperatures below T_K. Generally, an elastic constant c is related to the linear susceptibility with respect to Δ, as shown by the formula [14]:

$$c = c_0/(1 + g\chi_\Delta) \tag{7}$$

where c_0 is the elastic constant of the system, where there are no interactions between the lattice and the electronic system, and g is the coupling constant between Δ and the strain field. We assume that c_0 is independent of the temperature. The value of g is unknown

experimentally as well as theoretically. In order to discuss the crystalline field effect on c, we treat the factor g as a kind of fitting parameter. The quantity χ_Δ is calculated as the second-order derivative of the mean-field free energy:

$$\chi_\Delta = -\frac{\partial^2 F}{\partial \Delta^2} = \frac{1}{D}\int dE \frac{\tilde{V}^2}{(\tilde{E}_f - \Delta - E)^3} f(E-\mu) + \frac{1}{D}\int dE \frac{\tilde{V}^2}{(\tilde{E}_f + \Delta - E)^3} f(E-\mu) \tag{8}$$

where the Δ dependences of the band edges are neglected in the derivatives because their effect is exponentially small.

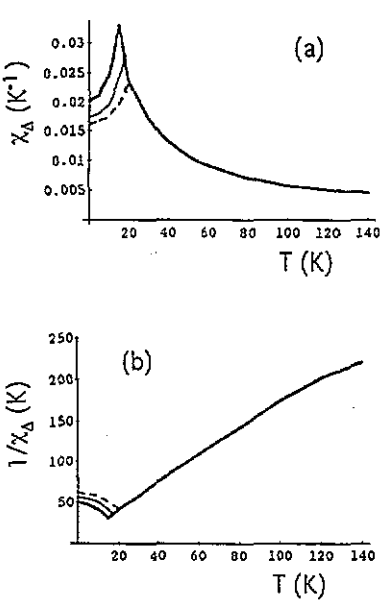

Figure 2. Temperature dependences of the linear susceptibility: (*a*) χ_Δ and (*b*) $1/\chi_\Delta$. The parameters and notations are the same as in figure 1.

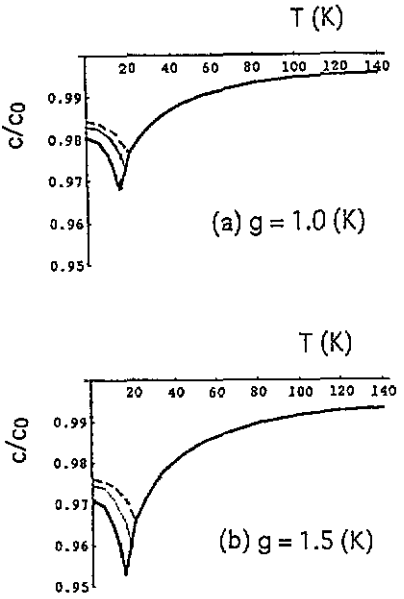

Figure 3. Temperature dependences of the elastic constant c/c_0 for (*a*) $g = 1.0$ K and (*b*) $g = 1.5$ K. The parameters and notations are the same as in figure 1.

Figure 2(*a*) displays the temperature dependence of χ_Δ. The inverse of χ_Δ is shown in figure 2(*b*). There is a peak at low temperatures. When $\Delta = 0$, χ_Δ is identical with the magnetic susceptibility. The peak of the curve for $\Delta = 0$ has been found in a previous paper [12] and is the effect of the fourfold degeneracy. When Δ is finite, the peak moves to a lower temperature and the peak height becomes taller. This is the crystalline field effect. The curve at high temperatures is less affected by Δ. The inverse of the susceptibility at high temperatures is almost linear against T, showing Pauli behaviour. The increase of χ_Δ at temperatures below the peak is well explained by the analytical expression

$$\chi_\Delta = (T_K^2(0) + 2\Delta^2)/T_K(\Delta)T_K^2(0) \tag{9}$$

at $T = 0$.

We show the temperature dependences of c/c_0, assuming several values for g; we plot two sets of curves for $g = 1$ K and 1.5 K, in figures 3(*a*) and (*b*), respectively. The elastic constant decreases from much higher to lower temperatures than T_K. This is the effect of the valence fluctuation. There is a downward dip around 15–20 K, which is the result of

the combination of the fourfold orbital degeneracy and the crystalline field splitting. The larger Δ gives rise to the larger dip.

We now look at the relationship with actual compounds. The elastic constant C_{33} of $CeCu_6$ [10] has a dip at about 10 K. This might be due to a Δ larger than the Kondo temperature 4 K. The constant $(C_{11} - C_{12})/2$ of CeTe [11] shows the apparent dip at about 15 K. The ground-state levels of Ce ions in CeTe split into the Γ_7 Kramers doublet and the Γ_8 quartet states. The Γ_7 states are the ground states. There is a splitting $2\Delta = 30$ K between Γ_7 and Γ_8 states. Figure 1 of [11] shows that the decrease of $(C_{11} - C_{12})/2$ constant from that of the ideal system without the electron–crystalline-field coupling is about 4% of the magnitude. This behaviour is simulated well by the thin curve in figure 3(*b*). The value $g = 1.5$ K gives us useful information on the coupling strength of heavy fermions against the crystalline field in CeTe.

5. Concluding remarks

We have solved the mean-field equations of the Anderson lattice model with crystalline field splitting 2Δ and calculated the linear susceptibility with respect to Δ. Next, we simulated the temperature dependence of the elastic constant, which is derived by the coupling of the electronic system to the crystalline field. We have discussed the relationship to experiment.

The downward dip of c exists even if there is no crystalline field splitting, i.e. $\Delta = 0$. The finite Δ results in a motion of the dip to lower temperatures. The magnitude of the dip becomes larger at the same time. The temperature where the dip is present would finally be determined by a combination of the high degeneracy of the 4f orbitals and the splitting width.

An interesting future problem would be a study of magnetic field effects. Further removal of the degeneracy by the magnetic field will give rise to more structures in the elastic constants. A theory of these effects would give helpful information for experimental studies.

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