Elastic anomaly of heavy fermion systems
in a crystalline field

Short title: Elastic anomaly of heavy fermion systems

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Abstract

An elastic anomaly, observed in the heavy fermi liquid state of Ce alloys (for example, CeCu₆ and CeTe), is analyzed by using the infinite-U Anderson lattice model. The 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

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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 nonzero value of the mean field of the slave bosons persists over the temperatures much higher than the Kondo temperature. This is the effect of the constant electron number. Next, the linear susceptibility with respect to $\Delta$ is calculated in order to obtain the renormalized elastic constant. The resulting temperature dependence of the constant shows the downward dip. We point out the relation of our finding with the experimental data.
§1. Introduction

Generally speaking, 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 the $j = 5/2$ 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. Secondly, 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 one of recent topics 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 renomalization [3]. The result shows the variation of the Kondo temperature $T_K$ with the temper-
ature. It comes from the change of the impurity scattering channel. For example, we imagine the case where there are three Kramers doublets with the splitting $\Delta_1$ and $\Delta_2$. In the temperature range $T < \Delta_1$, the three levels act independently in the scattering. But, in $\Delta_1 < T < \Delta_2$, the lower two levels behave as if they are one level of the degeneracy four, because thermal fluctuation has a larger width than $\Delta_1$. In the same way, in $T > \Delta_2$, all the three levels look as a level sixfold degenerate. It has been discussed that the above features are exhibited in temperature dependence of the magnetic susceptibility.

Experimentally, the heavy fermion systems show elastic anomalies related with valence instabilities in 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$_2$Si$_2$ [6]. The temperature dependences of the elastic constants have been theoretically fitted quantitatively well [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 $\alpha$- and $\gamma$-Ce. These elastic anomalies are related with the overall instability of the valence mainly due to the change of the mixing interaction. They are observed in the wide tem-
perature range $0 < T < T_K$ and 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 10K. This might be due to the splitting larger than the Kondo temperature 4K. The constant $(C_{11} - C_{12})/2$ of CeTe [11] shows the apparent dip at about 15K. The ground state levels of Ce ions split into the $\Gamma_7$ Kramers doublet and the $\Gamma_8$ quartet states. The $\Gamma_7$ states are the ground states. There is the splitting 30K between $\Gamma_7$ and $\Gamma_8$ 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 of them, smaller than $T_K$, means the width of the crystalline field splitting. And, 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 §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 owing to the fact that the Fermi level is varied in order to keep the total electron number constant. This is shown in §3.

Secondly, we calculate the linear susceptibility with respect to the crystalline field splitting. At lower temperatures, the susceptibility has a structure related with 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 is originated 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 kind of a parameter. We show temperature dependences of elastic constant for several choices of the coupling. We will discuss relevant parameters for the elastic
anomaly, i.e., the downward dip which is observed in the constant \((C_{11} - C_{12})/2\) of CeTe [11]. The susceptibility and elastic constant are reported and discussed in §4.

Finally, the paper is closed with several remarks in §5.

§2. Formalism

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

\[
H = \sum_i [(E_f - \Delta) \sum_{l = 1,2} f_{i,l}^\dagger f_{i,l} + (E_f + \Delta) \sum_{l = 3,4} f_{i,l}^\dagger f_{i,l}] + \sum_{\vec{k}, l = 1-4} \varepsilon_{\vec{k}} c_{\vec{k}, l}^\dagger c_{\vec{k}, l} + V \sum_{i, l = 1-4} (f_{i,l}^\dagger c_{i,l} b_i + b_{i}^\dagger c_{i,l}^\dagger f_{i,l}) + \sum_i \lambda_i (\sum_{l = 1-4} f_{i,l}^\dagger f_{i,l} + b_{i}^\dagger b_{i} - 1),
\]

where \(f_{i,l}\) is an annihilation operator of the f-electron of the \(l\)-th orbital at the \(i\)-th site, \(c_{\vec{k}, l}\) is an operator of the conduction electron with the wave number \(\vec{k}\), 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 \(E_f + \Delta\). The crystalline field splitting \(2\Delta\) 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, \(-D < \varepsilon_{\vec{k}} < (N - 1)D\), 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 Langrange multiplier field \( \lambda_{i} \).

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:

(1) the constraint condition,

\[
\frac{1}{2D} \int dE \frac{\tilde{V}^2}{(E_{\uparrow} - \Delta - E)^2} f(E - \mu)
\]

\[
+ \frac{1}{2D} \int dE \frac{\tilde{V}^2}{(E_{\uparrow} + \Delta - E)^2} f(E - \mu) + r^2 = 1,
\]

(2) the self-consistency condition for \( r \),

\[
\frac{1}{2D} \int dE \frac{V^2}{E - E_{\uparrow} + \Delta} f(E - \mu)
\]

\[
+ \frac{1}{2D} \int dE \frac{V^2}{E - E_{\uparrow} - \Delta} f(E - \mu) + \lambda = 0,
\]
and (3) the conservation condition of electron number $n_{el}$,

$$\frac{1}{2D} \int dE [1 + \frac{\tilde{V}^2}{(\tilde{E}_f - \Delta - E)^2}] f(E - \mu)$$

$$+ \frac{1}{2D} \int dE [1 + \frac{\tilde{V}^2}{(\tilde{E}_f + \Delta - E)^2}] f(E - \mu) = n_{el},$$

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 all the energy region of the bands. The three equations are solved numerically for the variables, $r$, $\lambda$, and the Fermi level $\mu$. In addition, the values at $T = 0$ can be obtained analytically.

§3. Solution

Equations (2), (3), and (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 $\Delta$ is increased from 0 but the magnitude is taken below the Kondo temperature $T_K = \tilde{E}_f - \mu$.

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 $\Delta = 0, 15, \text{ and } 30 K$ are shown by the dashed, thin, and thick lines, 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 \( \mu \) to keep the total electron number constant. This effect has been reported previously [12,13]. According to the increase of \( n_f \), \( \tilde{E}_f \) decreases, which means the 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 \( 2\Delta \). 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 \( T = 0 \). Small crystalline field splitting is assumed for the derivation. The results are

\[
T_K(\Delta) \equiv \tilde{E}_f - \mu = \sqrt{T_K^2(0) + \Delta^2} \tag{5}
\]

and

\[
r^2 \approx \frac{DT_K^2(0)}{V^2 T_K(\Delta)}, \tag{6}
\]

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 \( \Delta \) increases.
§4. Elastic anomaly in low temperatures

We shall discuss the change of elastic property of heavy fermions due to the crystalline field splitting in the low temperature below $T_K$. Generally, an elastic constant $c$ is related with the linear susceptibility with respect to $\Delta$, as shown by the formula [14]:

$$c = \frac{c_0}{1 + g \chi_\Delta},$$

where $c_0$ is the elastic constant of the system where there is not interactions between the lattice and the electronic system, and $g$ is the coupling constant between $\Delta$ 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 parameters. The quantity $\chi_\Delta$ is calculated as the second order derivative of the mean field free energy:

$$\chi_\Delta = \frac{\partial^2 F}{\partial \Delta^2},$$

$$= \frac{1}{D} \int dE \frac{\tilde{V}^2}{(E_f - \Delta - E)^3} f(E - \mu)$$

$$+ \frac{1}{D} \int dE \frac{\tilde{V}^2}{(E_f + \Delta - E)^3} f(E - \mu),$$

where the $\Delta$ dependences of the band edges are neglected in the derivatives because their effect is exponentially small.
Figure 2 (a) displays the temperature dependence of $\chi_\Delta$. The inverse of $\chi_\Delta$ is shown in Fig. 2(b). There is a peak at the low temperature. When $\Delta = 0$, $\chi_\Delta$ is identical with the magnetic susceptibility. The peak of the curve for $\Delta = 0$ has been found in the previous paper [12], and is the effect of the fourfold degeneracy. When $\Delta$ is finite, the peak moves to the lower temperature and the peak height becomes taller. This is the crystalline field effect. The curve at high temperatures is less affected by $\Delta$. The inverse of the susceptibility in high temperatures is almost linear against $T$, showing the Pauli behavior. The increase of $\chi_\Delta$ in the temperatures below the peak is well explained by the analytical expression:

$$\chi_\Delta = \frac{T_K^2(0) + 2\Delta^2}{T_K(\Delta)T_K^2(0)},$$

at $T = 0$.

We show the temperature dependences of $c/c_0$, assuming several values for $g$. We plot two sets of curves for $g = 1K$ and 1.5K, in Figs. 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 $\Delta$ gives rise to the larger dip.
We shall look at relations with actual compounds. The elastic constant $C_{33}$ of CeCu$_6$ [10] has a dip at about 10K. This might be due to $\Delta$ larger than the Kondo temperature 4K. The constant $(C_{11} - C_{12})/2$ of CeTe [11] shows the apparent dip at about 15K. The ground state levels of Ce ions in CeTe split into the $\Gamma_7$ Kramers doublet and the $\Gamma_8$ quartet states. The $\Gamma_7$ states are the ground states. There is the splitting $2\Delta = 30$K between $\Gamma_7$ and $\Gamma_8$ states. Fig. 1 of ref. [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 behavior is simulated well by the thin curve in Fig. 3(b). The value $g = 1.5$K gives us a 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 $2\Delta$, and calculated the linear susceptibility with respect to $\Delta$. Next, we have 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 relation with experiments.

The downward dip of $c$ exists even if there is no crystalline field splitting, i.e., $\Delta = 0$. The finite $\Delta$ results in the 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 be finally determined by
the combination of the high degeneracy of the 4f orbitals and the splitting
width.

One of the interesting future problems would be the magnetic field effects.
Further removal of the degeneracy by the magnetic field will give rise to
more structures in the elastic constants. The theory of the effects would be
a helpful information for experimental studies.

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Figure Captions

Fig. 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 dashed, thin, and heavy lines are for $\Delta = 0$, 15, and 30K, respectively.

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

Fig. 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 Fig. 1.