The elastic anomaly of heavy-fermion systems in a crystalline field: II. A realistic model for cubic crystals

1994 J. Phys.: Condens. Matter 6 977

(http://iopscience.iop.org/0953-8984/6/5/007)

J. Phys.: Condens. Matter 6 (1994) 977–984. Printed in the UK

The elastic anomaly of heavy-fermion systems in a crystalline field: II. A realistic model for cubic crystals

Kikuo Harigaya† and G A Gehring
Department of Physics, University of Sheffield, Sheffield S3 7RH, UK

Received 5 November 1993

Abstract. The elastic anomaly observed in the coherent Kondo state of Ce heavy-fermion compounds is analysed using the Anderson lattice model simulating the energy level scheme of the CeTe cubic crystal. The Γ_7 doublets and Γ_8 quartets of the 4f states are considered in the model. We solve the mean field equations to derive the temperature dependencies of elastic constants, using the random phase approximation for the interaction between the elastic strain and the crystalline field splitting. We compare the calculation with the $(c_{11} - c_{12})/2$ and c_{44} constants of CeTe. The presence of the downward dip and the observed overall temperature variations of the two constants are well described by the present theory. The origin of the dip is the coupling between the elastic strain and the splitting of the Γ_8 quartets.

1. Introduction

Heavy-fermion compounds show elastic anomalies at low temperatures. These are related to crystalline field structures, magnetic phase transitions, and so on. The main contribution from the crystalline fields is the peak or dip structures in the temperature dependencies of the elastic constants. Most of the crystalline field splittings are larger than the Kondo temperature and the anomalies occur at higher temperatures than those of the coherent Kondo state. For example, the elastic constant c_{33} of $CeCu_6$ [1] has a dip at about 10 K. This is due to the splitting being larger than the Kondo temperature 4 K. However, a few compounds have splittings comparable to or smaller than the Kondo temperature. These anomalies occur in the coherent Kondo state. The constants $(c_{11} - c_{12})/2$ and c_{44} of the CeTe cubic crystal [2] show an apparent dip at about 15 K. The $j = \frac{5}{2}$ levels of the Ce ions split into the Γ_7 Kramers doublet and the Γ_8 quartet states. The Γ_7 states are the ground states. The splitting between Γ_7 and Γ_8 states is 30 K. This is the origin of the dip. Similarly, the c_{44} constant of the alloy $Ce_{0.5}La_{0.5}B_6$ shows a dip at about 0.2 K, and this temperature is lower than the Kondo temperature of this alloy [3].

The main purpose of the present paper is to develop a theoretical description of the elastic anomaly by using a microscopic model. We use the Anderson lattice model, simulating the energy level scheme of CeTe. The Γ_7 doublets and Γ_8 quartets are considered for the cubic crystal. We solve the mean field equations and assume that a strain field couples linearly with the splitting of the Γ_8 states. We calculate the linear susceptibility with respect to the strain field. Then, we derive the temperature dependence of the elastic constant, using the random phase approximation (RPA) as in [4], which includes the elastic susceptibility. We

† Permanent address: Fundamental Physics Section, Physical Science Division, Electrotechnical Laboratory, Umezono 1-1-4, Tsukuba, Ibaraki 305, Japan; E-mail address: harigaya@etl.go.jp.

0953-8984/94/050977+08$07.50

compare the calculation with the $(c_{11}-c_{12})/2$ and c_{44} constants of CeTe. The observed overall temperature variations of the two constants are well described by the present theory including the coupling between the elastic strain and the splitting of the Γ_8 quartets.

In our previous paper [4], the case of the atomic f level with two doublets was considered and the elastic anomaly was discussed. In this paper, the realistic case for CeTe is studied for a model with one doublet and one quartet state. The calculated temperature variation of the elastic constant is compared with the experimental data.

We explain the model in section 2. We report the solution in section 3 and the elastic properties in section 4. We summarize our findings and give a discussion in section 5.

2. Formalism

We formalize the infinite-U Anderson lattice model simulating the crystalline field structures of CeTe [2] using the slave boson method. The model has the following form:

$$H=\sum_i\Bigg(E_f\sum_{l=1,2}+(E_f+\Delta-\delta)\sum_{l=3,4}+(E_f+\Delta+\delta)\sum_{l=5,6}\Bigg)f_{i,l}^\dagger f_{i,l}+\sum_{k,l=1-6}\varepsilon_k c_{k,l}^\dagger c_{k,l}$$
$$+V\sum_{i,l=1-6}(f_{i,l}^\dagger c_{i,l}b_i+b_i^\dagger c_{i,l}^\dagger f_{i,l})+\sum_i\lambda_i\Bigg(\sum_{l=1-6}f_{i,l}^\dagger f_{i,l}+b_i^\dagger b_i-1\Bigg) \qquad (1)$$

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 the wave number k, and b_i is an operator of the slave boson that indicates the unoccupied state at the f orbital. The atomic energy of the first and second orbitals of f electrons is E_f; that of the third and fourth orbitals is $E_f+\Delta-\delta$; and that of the third and fourth orbitals is $E_f+\Delta+\delta$. The two crystalline field splitting parameters Δ and δ are considered in the model. The first one Δ is the splitting between the ground state Γ_7 doublet and the excited Γ_8 quartet states of f electrons. The second splitting δ is due to the lattice distortion from the cubic symmetry. It is assumed that δ couples with a strain field ε linearly: $\delta=\eta\varepsilon$, where η is the coupling constant. For the conduction electrons, the same quantum number is assumed as for 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 $N=6$ 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 $\sum_{l=1-6}f_{i,l}^\dagger f_{i,l}+b_i^\dagger b_i=1$ 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 [4]:

(i) the constraint condition

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

(ii) the self-consistency condition for r

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

and

(iii) the conservation condition of electron number n_{el}

$$\frac{1}{3D}\int dE\left(1+\frac{\tilde{V}^2}{(\tilde{E}_f+\Delta+\delta-E)^2}\right)f(E-\mu)+\frac{1}{3D}\int dE\left(1+\frac{\tilde{V}^2}{(\tilde{E}_f+\Delta-\delta-E)^2}\right)$$
$$\times f(E-\mu)+\frac{1}{3D}\int dE\left(1+\frac{\tilde{V}^2}{(\tilde{E}_f-E)^2}\right)f(E-\mu)=n_{el} \tag{4}$$

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

3. Solution

Equations (2), (3) and (4) are solved numerically for the parameters $D=5\times10^4$ K, $V=7500$ K, $E_f=-10^4$ K, and $n_{el}=1.9$ as the typical values. We take the splitting parameter $\Delta=30$ K. The small value for Δ has been revealed in experiment [5]. We consider the limit $\delta\to0$ because the splitting from the strain is so small as to be negligible. As we will see later, Δ is smaller than the Kondo temperature $T_K=\tilde{E}_f-\mu$ at $T=0$ K.

Figure 1 shows the temperature dependencies of the parameters. Figure 1(a), (b), and (c) shows the variations of $\tilde{E}_f$, T_K, and the number n_f of f electrons per site, respectively. 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 (about 40 K) at $T=0$. This is the effect of the change of the Fermi level μ, keeping the total electron number constant. This effect has been reported previously [6, 7]. Corresponding to the increase of n_f, $\tilde{E}_f$ decreases, which means a reduced itinerancy of f electrons owing to the increase of n_f. 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 an increased value of n_f when the crystalline field is switched on. Also, $\tilde{E}_f$ decreases and T_K increases, due to the crystalline field. A similar dependence on Δ has been reported in our previous paper [4]. The Kondo temperature $T_K(\Delta)$ (at $T=0$) as a function of Δ satisfies the equation $[T_K(\Delta)+\Delta]^2T_K^2(\Delta)=T_K^3(0)$, where $T_K(0)=D\exp[-D(\mu-E_f)/V^2]$ is the Kondo temperature for $\Delta=0$. Starting from this analytic expression, we could verify the low temperature variations of the parameters by using the expansion with respect to $\Delta/T_K(0)$, assuming small Δ.

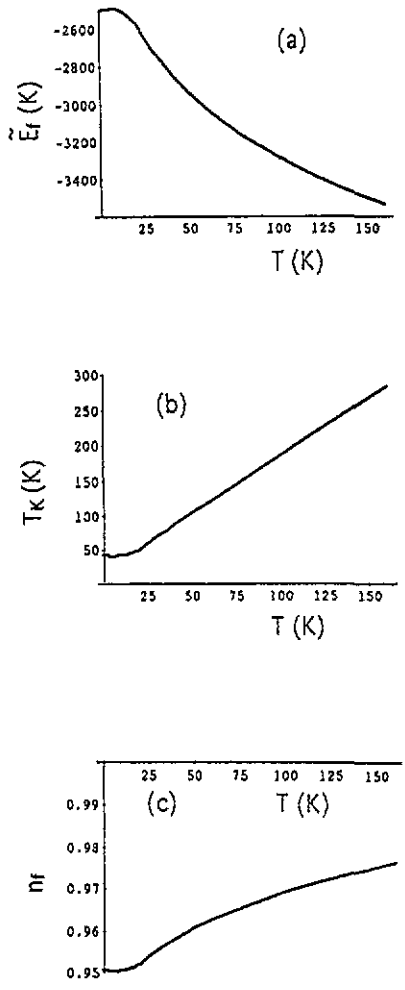

Figure 1. Temperature dependencies of the mean field solution: (a) $\tilde{E}_f$, (b) T_K, and (c) n_f. Parameters are $D = 5 \times 10^4$ K, $V = 7500$ K, $E_f = -10^4$ K, $n_{el} = 1.9$, and $\Delta = 30$ K.

4. Elastic anomaly at low temperatures

We shall discuss the change of elastic properties of heavy fermions due to the crystalline field splitting at low temperature below T_K. We shall calculate an elastic constant c by the RPA-like formula [4] analogous to the plasmon excitation theory. The constant c is related to the linear susceptibility with respect to δ, as shown below:

$$c = \frac{c_0}{1 + g\chi_\delta} \tag{5}$$

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. These constants are related as $g = c_0\eta^2$, so g is positive. An analogous formula has been used before [8] but in the

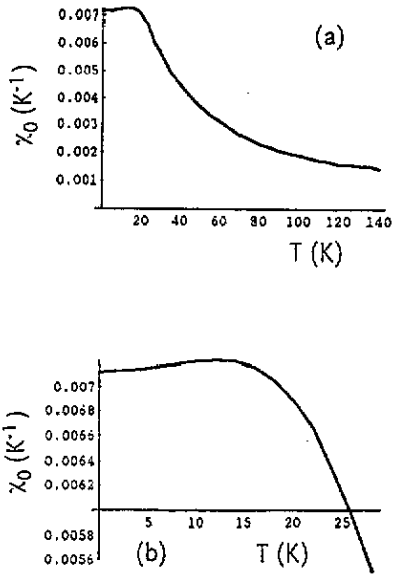

Figure 2. Temperature dependencies of the linear susceptibility χ_0. The parameters are the same as in figure 1. (b) shows the structures at low temperature from (a).

linear response theory. 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:

$$\chi_\delta = -\frac{\partial^2 F}{\partial \delta^2} = \frac{2}{3D}\int \mathrm{d}E \, \frac{\tilde{V}^2}{(\tilde{E}_f + \Delta - \delta - E)^3} f(E-\mu) + \frac{2}{3D}\int \mathrm{d}E \, \frac{\tilde{V}^2}{(\tilde{E}_f + \Delta + \delta - E)^3} f(E-\mu) \tag{6}$$

where the δ dependencies of the band edges are neglected in the derivatives because their effect is exponentially small. In the actual calculation, we take the limit $\delta \to 0$, because our problem is the elastic property at the equilibrium of the cubic lattice where there is no splitting δ.

Figure 2 displays the temperature dependence of $\chi_0 \equiv \lim_{\delta \to 0} \chi_\delta$. Figure 2(a) shows the variation over a wide temperature range, and figure 2(b) shows the detailed structure at low temperature. There is a peak around $T = 15$ K. This is owing to the large degree of freedom for electrons and the crystalline field Δ. The position of the peak would depend on parameters, but here the position agrees with that of the $(c_{11} - c_{12})/2$ constant of CeTe [2]. The appearance of the peak has been discussed in our previous paper [4]. The susceptibility at high temperature is nearly inversely proportional to T, showing Pauli paramagnetic behaviour. The value of χ_0 at $T = 0$ is analytically expressed as

$$\chi_0 = \frac{2T_K(\Delta)}{[T_K(\Delta) + \Delta][3T_K(\Delta) + \Delta]} \tag{7}$$

using the Kondo temperature.

Now, we compare the calculated c/c_0 with the experiments. We plot the temperature dependencies c/c_0, obtained from the experimental data for CeTe [2], shown by the dots in figure 3(a) and (b) for the $(c_{11}-c_{12})/2$ and c_{44} constants, respectively. The experimental c_0 depends on the temperature. We used the linear dependence given in [2] for the $(c_{11}-c_{12})/2$ mode. The quantity c_0 becomes softer as the temperature increases. However, we cannot use the experimental c_0 for the c_{44} mode [2] in order to compare with the theory. We rather use the increasing linear function $c_0(T) = 0.696 + 9.89 \times 10^{-5}T$ (10^{11} erg cm^{-3}). It seems strange that the constant becomes larger as T rises, but this does not mean that the crystal becomes harder for increasing T. Elastic constants can become harder as T increases, in fact, c_{44} becomes harder, and c_{11} and $(c_{11}-c_{12})/2$ become softer in CeTe. In the two figures, the calculated c/c_0 is shown by the curve for $g = 6.9$ K. The elastic constant decreases from much higher to lower temperatures than T_K. The decrease is almost proportional to $1/T$. The agreement is fairly good. The decrease is the effect of the valence fluctuation. There is a downward dip around 15–20 K and the position also agrees with the experiments. The overall temperature dependencies are well explained by the same g for the two constants. As both c_0 and η are different for the two modes, this result should be regarded as a coincidence.

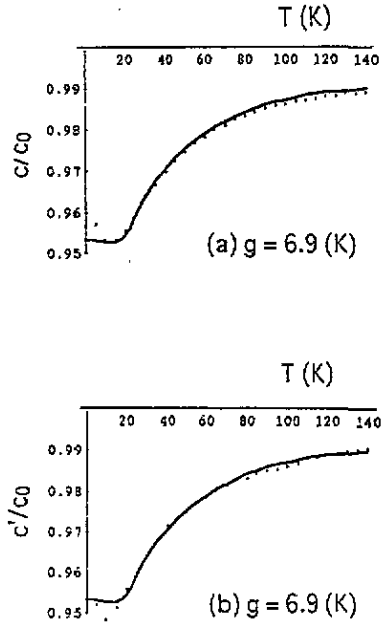

Figure 3. Temperature dependencies of the elastic constant c/c_0 for $g = 6.9$ K. The parameters are the same as in figure 1. In (a), the calculation is compared with the $(c_{11}-c_{12})/2$ constant of CeTe. In (b), the comparison with the c_{44} constant is made. Experimental data are shown by the dots.

5. Summary and discussion

We have solved the mean field equations of the Anderson lattice model with the crystalline field splitting between Γ_7 doublets and Γ_8 quartets. It has been assumed that the strain field couples linearly with the splitting of the Γ_8 states. We have calculated the linear susceptibility with respect to the strain field. Next, we have derived the temperature dependence of the elastic constant, using an RPA-like expression that includes the elastic susceptibility. We have compared the calculation with the $(c_{11}-c_{12})/2$ and c_{44} constants of CeTe. The observed overall temperature variations of the two constants are well described by the present theory including the coupling between the elastic strain and the splitting of the Γ_8 quartets. We believe that the presence of the peak in χ_0 is not an artifact of the mean field theory. In fact, the magnetic susceptibility of the exact solution of the single site system has a peak when the number of degrees of freedom is larger than two.

In the actual compound, V is anisotropic, i.e., it has an angle dependence in the momentum space: $V = V(\theta, \phi)$. The mean field equations (2), (3) and (4) change only in one point: the angle average

$$\int \frac{\mathrm{d}(\cos\theta)\,\mathrm{d}\phi}{4\pi}\, V^2(\theta,\phi) \tag{8}$$

appears in the equations. This angle average can be absorbed in the present formalism by regarding the angle averaged $\sqrt{V^2}$ as the isotropic V of (1). Thus, the parameter value V used in this paper should be interpreted as an averaged one.

In [3], the alloy system $Ce_{0.5}La_{0.5}B_6$ has a downward dip in the temperature dependence of the c_{44} constant. The variation is very similar to that in figure 3. The same mechanism of the elastic anomaly discussed in this paper would also work in this alloy system. The elastic anomaly in magnetic alloys could be treated by a microscopic theory using the coherent potential approximation applied to the Anderson alloy system [9]. This calculation would be an interesting extension of this paper.

By applying the magnetic field, the atomic energy levels of Ce will split further. This will result in more structure in the temperature variations of the elastic constants. Experimental as well as theoretical information would be useful for detailed understanding of the electronic properties of heavy-fermion systems.

Acknowledgments

The authors thank Professor T Goto for sending them copies of his papers and the fruitful discussion. They also thank Dr H Matsui for the useful discussion. This work was done while one of the authors (KH) was staying at the University of Sheffield on leave from the Electrotechnical Laboratory, Japan. He acknowledges financial support for the stay obtained from the SERC and the hospitality that he obtained from the University.

References

[1] Goto T, Suzuki T, Ohe Y, Fujimura T, Sakutsume S, Onuki Y and Komatsubara T 1988 *J. Phys. Soc. Japan* **57** 2612

[2] Matsui H, Goto T, Tamaki A, Fujimura T, Suzuki T and Kasuya T 1988 *J. Magn. Magn. Mater.* **76/77** 321

[3] Nakamura S, Goto T, Matsui H, Sakatsume S and Kunii S 1993 *Preprint*
[4] Harigaya K and Gehring G A 1993 *J. Phys.: Condens. Matter* **5** 5277
[5] Rossat-Mignod J, Effantin J M, Burlet P, Chattopadhyay T, Regnault L P, Bartholin H, Vettier C, Vogt O, Ravot D and Achart J C 1985 *J. Magn. Magn. Mater.* **52** 111
[6] Evans S M M, Chung T and Gehring G A 1989 *J. Phys.: Condens. Matter* **1** 10473
[7] Harigaya K 1988 *Master Thesis* University of Tokyo
[8] Levy P 1973 *J. Phys. C: Solid State Phys.* **6** 3545
[9] Harigaya K 1990 *J. Phys.: Condens. Matter* **2** 4623