Theory of elastic properties of Sr_2RuO_4 at the superconducting transition temperature

M. B. Walker and Pedro Contreras
Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7
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This paper gives details of a predicted splitting of the superconducting transition temperature in Sr_2RuO_4 as a function of applied uniaxial stress. We also give formulas for the discontinuities in certain thermodynamic properties such as the specific heat, the thermal expansion coefficients, and the elastic compliance coefficients at various superconducting transition temperatures.

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INTRODUCTION

Superconductivity in Sr_2RuO_4 (Ref. 1) has a number of unusual properties which continue to attract interest and further study (Ref. 2 gives an excellent review). The crystal structure of this material is identical to that of the high-temperature cuprate superconductor (La, Sr)$_2$CuO$_4$, with ruthenium ions replacing the copper ions. Also the normal state of Sr_2RuO_4 displays Fermi-liquid behavior. The Cooper pairs are believed to form in a spin-triplet state,[3–5] and the superconducting parameter is believed to break time-reversal symmetry.[6] Studies aimed at elucidating the detailed nature of the superconducting pairing symmetry continue to be carried out (e.g. see Refs. 7, 8, 9, and 10). Recent experimental measurements[11] of the elastic constants of Sr_2RuO_4 as the temperature is lowered through the superconducting transition temperature give further interesting insights into the nature of its superconducting state. These elastic constant measurements stimulated our interest in developing a theoretical model for the changes occurring in the elastic constants at the superconducting transition temperature, and this interest then developed into the more general study of the elastic behavior of Sr_2RuO_4 under applied uniaxial stress which is reported here. As will be noted below, a full understanding of the elastic behavior of Sr_2RuO_4 in zero stress cannot be obtained without also understanding the behavior in the presence of nonzero uniaxial stress.

The use of the thermodynamic Ehrenfest relations and Ginzburg-Landau theories is a useful tool in the analysis of sound velocity measurements at a normal metal to superconducting state phase transition,[12–17] and such methods have recently been applied to Sr_2RuO_4.[18–20] The analysis of sound velocity measurements performed under conditions of zero externally applied stress in Sr_2RuO_4 requires special consideration,[21] however, because the derivative of the transition temperature with respect to uniaxial stress (one of the parameters occurring in the Ehrenfest relations) is not defined at the point of zero applied stress. This is because an external uniaxial stress will split the superconducting phase transition into two transitions. Under these conditions quantities such as the elastic constants will have discontinuities at each of the two transitions, and it is the sum of these two discontinuities that will that will be equal to the single discontinuity that occurs when the uniaxial stress tends to zero.

Superconductivity in the heavy-fermion metal UPt_3 (for a review, see Ref. 17) has features similar to those in Sr_2RuO_4. Both are thought to be characterized by two-component superconducting order parameters. Furthermore, the superconducting transition temperature in UPt_3 is split as a result of a symmetry-breaking antiferromagnetism which exists in UPt_3, in much the same way as the transition temperture in Sr_2RuO_4 might be expected to split in the presence of a symmetry-breaking stress. However in UPt_3 the symmetry-breaking antiferromagnetism is not under the control of the experimenter, and cannot be made zero in the superconducting state. This means that some of the qualitative features discussed below have not been investigated in connection with UPt_3. Further complications arise in UPt_3 because the antiferromagnetic moment directions can take one of three different directions in the basal plane, and this gives rise to a domain structure which cannot be supressed.[22] The presence of this domain structure in UPt_3 very much limits the experimental information one can obtain on symmetry-breaking effects in this material, since each domain has its own direction of the symmetry-breaking field, and measurements on a macroscopic sample thus give average properties of the three domains. In Sr_2RuO_4 on the other hand, the symmetry-breaking field (uniaxial stress) is under the control of the experimenter, with the states of zero symmetry-breaking stress, and of a single direction of uniaxial stress, being easily attainable. Thus, it would appear that there would be significant advantages to the use of Sr_2RuO_4 as a material for a detailed study of symmetry-breaking effects in superconductivity described by a two-component order parameter. One relative disadvantage of Sr_2RuO_4 (discussed below) is that as a result of its tetragonal symmetry (versus hexagonal for UPt_3) there are a greater number of independent parameters in the Ginzburg-Landau model that will have to be determined from experimental measurements.

EHRENFEST RELATIONS AND GINZBURG-LANDAU MODEL

Ehrenfest relations have often been found to be useful in analyzing the changes in sound velocity occurring at a second-order phase transition.[12] Consider a second-order phase transition to a superconductor state where the transition temperature is, in principle, known as a function of an externally applied stress σ_i ($i=1, \ldots 6$ using Voigt notation) i.e., given $T_c = T_c(\sigma_i)$. Then, from the fact that the entropy and the state of strain are continuous at a second-order phase transition, the Ehrenfest relations

$$\Delta\alpha_i = -\Delta C_\sigma \frac{\partial \ln T_c}{\partial \sigma_i}, \quad \Delta s_{ij} = -\Delta\alpha_i \frac{\partial T_c}{\partial \sigma_j} \tag{1}$$

can be derived (e.g., see Ref. 23). Here $\alpha_i = (\partial e_i/\partial T)_\sigma$ is the coefficient of thermal expansion, with e_i being the ith component of the strain. C_σ is the specific heat at constant stress, and s_{ij} is the elastic compliance matrix. Also, for any quantity Q, $\Delta Q = Q(T_c + 0^+) - Q(T_c - 0^+)$, where 0^+ is a positive infinitesimal, is the discontinuity in Q at $T = T_c$. Combining Eqs. (1) and (2) gives

$$\Delta s_{ij} = \frac{\Delta C_\sigma}{T_c} \frac{\partial T_c}{\partial \sigma_i} \frac{\partial T_c}{\partial \sigma_j}. \tag{2}$$

Because (as shown below) the superconducting transition temperature in Sr_2RuO_4 is split into two transitions by a uniaxial stress, these relations cannot be directly applied to the transition occurring at zero stress. Thus the phase diagram of Sr_2RuO_4 in nonzero uniaxial stress (which is of interest in its own right) will have to be investigated.

Similarly, if $T_c = T_c(p)$ is given as a function of hydrostatic pressure, the relations

$$\Delta\beta = \Delta C_p \frac{\partial \ln T_c}{\partial p}, \quad \Delta K = \frac{\Delta\beta}{V} \frac{\partial T_c}{\partial p} \tag{3}$$

can be derived. Here $\beta = (\partial V/\partial T)_p$ and $K = -V^{-1}(\partial V/\partial p)_T$. Also, from Eq. (4)

$$\Delta K = \frac{\Delta C_p}{T_c V} \left(\frac{\partial T_c}{\partial p} \right)^2. \tag{4}$$

The application of these relations to Sr_2RuO_4 is relatively straight forward, as there is no splitting of T_c with the application of hydrostatic pressure.

A more detailed description of the changes in thermodynamic properties occurring at a superconducting phase transition can be obtained in terms of a Ginzburg-Landau model for superconductivity. The order parameter describing superconductivity in Sr_2RuO_4 must transform according to one of the irreducible representations of the tetragonal point group D_{4h}. There are eight distinct one-dimensional representations of D_{4h} and two two-dimensional representations. Suppose that the superconductivity is described by an order parameter Ψ which transforms according to one of the eight one-dimensional representations. Then the terms in the Ginzburg-Landau free energy which are linear in stress and quadratic in the superconductor order parameter have the form

$$G_{int} = [a(\sigma_{xx} + \sigma_{yy}) + b\sigma_{zz}]|\Psi|^2. \tag{5}$$

It is these terms that give rise to discontinuities in the elastic constants measured in sound velocity measurements.[12] Note, however, that for reasons of symmetry, there is no coupling linear in σ_{xy}, which is the coupling that would give rise to a discontinuity in the elastic compliance constant s_{66} (and also in c_{66}). Hence s_{66} and c_{66} are continuous at T_c in this model, which assumes a one-dimensional order parameter. This is in contradiction to the experimental results[11] on Sr_2RuO_4, which show a discontinuity in c_{66}. From this we conclude, from sound velocity measurements only, that none of the eight one-dimensional irreducible representations can give an appropriate description of superconductivity in Sr_2RuO_4.

Superconductivity in Sr_2RuO_4 must therefore be described by an order parameter transforming as one of the two dimensional irreducible representation E_{2g} or E_{2u}. There is evidence that the E_{2u}, spin-triplet state is the correct choice (e.g., see Ref. 2), and this is what we will assume here. In fact however, the assumption of an E_{2g} spin-singlet state would give an identical description so that the sound velocity measurements analyzed in terms of the model presented here can not distinguish between E_{2g} and E_{2u} representations.

The model is specified by giving the Gibbs free energy associated with the two-component order parameter (Ψ_x, Ψ_y) transforming like the E_{2u} representation of the point group D_{4h}, which is

$$\begin{aligned} G = {} & G_0(T) + \alpha'(T - T_{c0})(|\Psi_x|^2 + |\Psi_y|^2) \\ & + \frac{b_1}{4}(|\Psi_x|^2 + |\Psi_y|^2)^2 + b_2|\Psi_x|^2|\Psi_y|^2 \\ & + \frac{b_3}{2}(\Psi_x^2\Psi_y^{*2} + \Psi_y^2\Psi_x^{*2}) - \frac{1}{2}s_{ij}\sigma_i\sigma_j + \sigma_i L_i + \sigma_i d_{ij} E_j . \end{aligned} \tag{6}$$

The nonzero components of E_i are $E_1 = |\Psi_x|^2$, $E_2 = |\Psi_y|^2$, and $E_6 = \Psi_x^*\Psi_y + \Psi_x\Psi_y^*$. The elastic compliance matrix s_{ij} and the matrix d_{ij} have the same symmetry properties, appropriate for tetragonal symmetry. The parameters L_i describe thermal expansion, so that if the state of the system at $T = T_{c0}$ is taken to be the refence state corresponding to zero strain, then for temperatures sufficiently close to T_{c0}, $L_i \approx (T - T_{c0})L_i'$, where L_i' is independent of temperature. Because not all components of E_i can be nonzero, the nonzero independent components of d_{ij} that enter into the model are d_{11}, d_{12}, d_{31}, and d_{66}. Terms that are quadratic in both the superconducting order parameter and the applied stress have been omitted from the above free energy. Such terms would give an additional temperature dependence to the elastic constants below the critical temperature.[12] Given the large number of independent constants occuring in the associated sixth-rank tensor, it is not clear that the explicit inclusion of such terms would be productive, at least at this stage of the investigation.

An analysis of the above model for zero stress ($\sigma_i = 0$ for all i) shows that we must have $b_3 > 0$ and $\tilde{b} \equiv b_3 - b_2 > 0$ for the reversal symmetry state $(\Psi_x, \Psi_y) = (1, \pm i)$ to have the lowest free energy, and $b \equiv b_1 + b_2 - b_3 > 0$ for the phase transition to be second order. This broken time-reversal-symmetry state is generally believed to be the state describing superconductivity in Sr_2RuO_4.[2]

Now consider, for example, the case of uniaxial compression along the a axis (only $\sigma_1 \neq 0$ and $\sigma_1 < 0$). Then the terms quadratic in the order parameter can be written in the form

$$\begin{aligned} G_{quad} = {} & \alpha'[T - T_{c+}(\sigma_1)]|\Psi_x|^2 + \alpha'[T - T_{cy}(\sigma_1)]|\Psi_y|^2 \\ & + \cdots, \end{aligned} \tag{7}$$

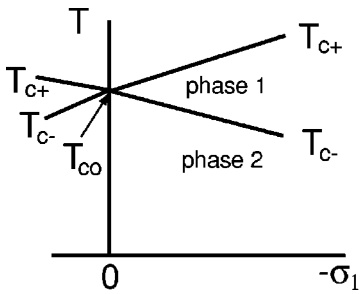

FIG. 1. Phase diagram showing the upper and lower superconducting transition temperatures, T_{c+} and T_{c-}, respectively, as functions of the compressive stress $-\sigma_1$ along the a axis.

where

$$T_{c+}(\sigma_1)=T_{c0}-\sigma_1 d_{11}/\alpha' \tag{8}$$

and $T_{cy}(\sigma_1)=T_{c0}-\sigma_1 d_{12}/\alpha'$. We will assume that $d_{11}-d_{12}>0$ so that $T_{c+}>T_{cy}$. (If $d_{11}-d_{12}<0$ an identical model can be obtained simple by interchanging the indices x and y.)

Since T_{c+} is the higher of the two critical temperatures, there will be an initial transition at T_{c+} when temperature is lowered, and just below T_{c+} only Ψ_x will be nonzero. As the temperature is lowered further, another phase transition occurs at a temperature called T_{c-} (which is different from T_{cy}), below which Ψ_y becomes nonzero. Thus, in the presence of a nonzero compressive stress σ_1, the order parameter has the form $(\Psi_x,\Psi_y)=\eta(1,\pm i\epsilon)$ where ϵ is real, and is zero in phase 1 (between T_{c+} and T_{c-}), and grows from $\epsilon=0$ to $\epsilon\approx 1$ as T is reduced below T_{c-} in phase 2. The temperature T_{c-} can be shown to be given in terms of the result

$$T_{c+}-T_{c-}=-\frac{d_{11}-d_{12}}{2\alpha'}\frac{\tilde{b}+b}{\tilde{b}}\sigma_1 . \tag{9}$$

The phase diagram just described is shown in Fig. 1. The splitting of the superconducting phase transition into two transitions by a symmetry-breaking stress, as shown here, is similar to the splitting of the superconducting phase transition in UPt_3 by a symmetry-breaking antiferromagnetic magnetization (e.g., see Ref. 13). However, the details are somewhat different as a result of the lower symmetry of Sr_2RuO_4 (tetragonal as opposed to hexagonal). Also, in the present case of Sr_2RuO_4 the symmetry-breaking stress can be reduced to zero, whereas this in not the case for UPt_3 where the symmetry-breaking magnetization density is not a parameter under the control of the experimenter.

Detailed calculations of the discontinuities in thermodynamic properties at T_{c+}, in the presence of a nonzero compressive stress σ_1, give

$$\Delta C_\sigma^+=-\frac{2T_{c+}\alpha'^2}{b_1},\quad \Delta\alpha_{i'+}=-\frac{2\alpha' d_{i'1}}{b_1}$$

$$\frac{\partial T_{c+}}{\partial\sigma_{i'}}=-\frac{d_{i'1}}{\alpha'},\quad \Delta s_{i'j'}^+=-\frac{2d_{i'1}d_{j'1}}{b_1}, \tag{10}$$

where a prime on an index (as in i' or j') indicates a Voigt index taking only the values 1, 2, or 3. These quantities can be shown to satisfy the Ehrenfest relations stated in Eqs. (1) and (2) above. Similarly detailed calculations of the discontinuities at T_{c-} yield the results

$$\begin{gathered}\Delta C_\sigma^-=-2T_{c-}\alpha'^2\frac{\tilde{b}}{bb_1},\\ \frac{\partial T_{c-}}{\partial\sigma_{i'}}=-\frac{1}{2\alpha'}\left(d_{i'+}-\frac{b}{\tilde{b}}d_{i'-}\right),\\ \Delta\alpha_{i'}^-=-\alpha'\frac{\tilde{b}}{bb_1}\left(d_{i'+}-\frac{b}{\tilde{b}}d_{i'-}\right),\\ \Delta s_{i'j'}^-=-\frac{\tilde{b}}{2bb_1}\left(d_{i'+}-\frac{b}{\tilde{b}}d_{i'-}\right)\left(d_{j'+}-\frac{b}{\tilde{b}}d_{j'-}\right),\end{gathered} \tag{11}$$

where $d_{i'\pm}=d_{i'1}\pm d_{i'2}$. These quantities satisfy the Ehrenfest relations for the discontinuities occurring at T_{c-}.

To find the discontinuities which occur at T_{c0} in the absence of stress, one can either take the sum of the discontinuities occurring at T_{c+} and T_{c-}, or calculate the discontinuities occurring at T_{c0} directly. Both give the same result, which is

$$\begin{gathered}\Delta C_\sigma^0=-\frac{2T_{c0}\alpha' 2}{b},\quad \Delta\alpha_{i'}^0=-\frac{\alpha' d_{i'+}}{b},\\ \Delta s_{i'j'}^0=-\frac{1}{2}\left(\frac{d_{i'+}d_{j'+}}{b}+\frac{d_{i'-}d_{j'-}}{\tilde{b}}\right).\end{gathered} \tag{12}$$

There is no reason to expect an Ehrenfest relation to hold at zero stress since the derivative of T_c with respect to stress is not defined at this point.

Now consider the phase diagram which occurs when only $\sigma_6\equiv\sigma_{xy}$ is non zero. The phase diagram is the same as in Fig. 1 except that σ_1 is replaced by σ_6, and T_{c+} and T_{c-} are now given by $T_{c+}(\sigma_6)=T_{c0}-\sigma_6 d_{66}/\alpha'$ and $T_{c-}(\sigma_6)=T_{c0}+\sigma_6 d_{66}b/(2b_3\alpha')$. The order parameter has the form $(\Psi_x,\Psi_y)=\eta(e^{i\varphi/2},e^{-i\varphi/2})$ where $\varphi=0$ in phase 1 of Fig. 1, and φ grows from 0 to approximately $\pi/2$ as the temperature is lowered below T_{c-} in phase 2. Also, the discontinuities at T_{c+} are

$$\begin{gathered}\Delta C_\sigma^+=-\frac{2T_{c+}\alpha'^2}{b'},\quad \Delta s_{66}^+=-\frac{2d_{66}^2}{b'},\\ \Delta\alpha_6^+=-\frac{2\alpha' d_{66}}{b'},\quad \frac{\partial T_{c+}}{\partial\sigma_6}=-\frac{d_{66}}{\alpha'}\end{gathered} \tag{13}$$

whereas the discontinuities at T_{c-} are

$$\Delta C_\sigma^-=-\frac{4T_{c-}\alpha'^2 b_3}{bb'},\quad \Delta s_{66}^-=-\frac{d_{66}^2 b}{b' b_3},$$

$$\Delta\alpha_6^- = +\frac{2\alpha' d_{66}}{b'}, \quad \frac{\partial T_{c-}}{\partial\sigma_6} = \frac{b d_{66}}{2b_3\alpha'} \tag{14}$$

where $b' = b_1 + b_2 + b_3 > 0$. The Ehrenfest relations of Eqs. (1) and (2) are obeyed at both T_{c+} and T_{c-}. The discontinuities at $\sigma_6 = 0$ and $T = T_{c0}$ are

$$\Delta C_\sigma^0 = -\frac{2T_{c0}\alpha'^2}{b}, \quad \Delta s_{66}^0 = -\frac{d_{66}^2}{b_3}, \quad \Delta\alpha_6^0 = 0. \tag{15}$$

The above work found it natural to make use of the 6×6 elastic compliance matrix S (matrix elements s_{ij}), because the phase diagram was determined as a function of the external stress (rather than a as function of strain). However sound velocity measurements are best interpreted in terms of the elastic stiffness matrix C (matrix elements c_{ij}), which is the inverse of the elastic compliance matrix. It is therefore useful to give a formula giving the discontinuities in the elastic stiffness matrix in terms of these of the elastic compliance matrix. To do this we write $C(T_c + 0^+) = C(T_c - 0^+) + \Delta C$, $S(T_c + 0^+) = S(T_c - 0^+) + \Delta S$, where 0^+ is positive infinitesimal, and make use of the fact that $C(T_c + 0^+)S(T_c + 0^+) = \mathbb{1}$, where $\mathbb{1}$ is the unit matrix. Then, to first order in the discontinuities,

$$\Delta C \approx -C\Delta S C, \tag{16}$$

where C can be taken to be the normal state elastic stiffness matrix. Thus, for example, at T_{c+} in the phase diagram of Fig. 1,

$$\Delta c_{11}^+ \approx \frac{2(c_{j1}d_{j1})^2}{b_1}, \tag{17}$$

From this equation it is clear that $\Delta c_{11}^+ > 0$.

Finally, it should be noted that the results of the discontinuities derived above satisfy the following inequalities at T_{c+}, T_{c-}, and T_{c0}. First, $\Delta C_\sigma < 0$. Second, the diagonal elements Δs_{11}, Δs_{22}, Δs_{33} and Δs_{66} are all negative. Third, the diagonal elements Δc_{11}, Δc_{22}, Δc_{33} and Δc_{66} are all positive. An important conclusion of this work is that the observation by Lupien *et al.*[11] of a discontinuity in the elastic constant c_{66} is evidence that the superconducting order parameter in Sr_2RuO_4 has two components.

The task of determining the magnitudes of the parameters in the Ginzburg-Landau model from experimental measurements performed on Sr_2RuO_4 will be somewhat more complicated in principle than the corresponding problem for UPt_3, because the number of independent parameters occurring for the case of tetragonal symmetry (Sr_2RuO_4) is greater than for the case of hexagonal symmetry (UPt_3). For example, there are three linearly independent parameters, b_1, b_2 and b_3 required to specify the terms of fourth order in the order parameter occurring in the free energy of Eq. (6) for Sr_2RuO_4, whereas only two independent parameters [called β_1 and β_2 (Refs. 13 and 14)] are required for UPt_3. In Refs. 13 and 14 it is noted that the ratio β_2/β_1 can be determined from the measurement of what we have called $\Delta C_\sigma^+/\Delta C_\sigma^-$. For Sr_2RuO_4, two independent ratios can be formed from the three independent b parameters, and these two independent ratios could be determined, for example, by experimentally determining the ratios $\Delta C_\sigma^+/\Delta C_\sigma^-$ in the presence of the stress σ_1 and also in the presence of the stress σ_6. In spite of this extra complexity, Sr_2RuO_4 might well be a better candidate than UPt_3 for a detailed experimental investigation of the effects of a symmetry-breaking field because, in the absence of externally applied symmetry-breaking stress, there is no symmetry-breaking field in Sr_2RuO_4. Thus the symmetry-breaking field is under the control of the experimenter. In UPt_3, on the other hand, there is always a symmetry-breaking field present due to the antiferromagnetism, and to make matters worse the magnetic moment directions in UPt_3 can take one of three different directions in the basal plane, which gives rise to a domain structure which cannot be supressed.[22] The presence of this domain structure in UPt_3 complicates the interpretation of experimental information on symmetry-breaking effects in this material.

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