

## Symmetry field breaking effects in $\text{Sr}_2\text{RuO}_4$

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In this work, after reviewing the theory of the elastic properties of  $\text{Sr}_2\text{RuO}_4$ , an extension suitable to explain the sound speed experiments of Lupien *et al.* [2] and Clifford *et al.* [3] is carried out. It is found that the discontinuity in the elastic constant  $C_{66}$  gives unambiguous experimental evidence that the  $\text{Sr}_2\text{RuO}_4$  superconducting order parameter  $\Psi$  has two components and shows a broken time-reversal symmetry state. A detailed study of the elastic behavior is performed by means of a phenomenological theory employing the Ginzburg-Landau formalism.

**Keywords:** Elastic properties; unconventional superconductors; time reversal symmetry; Ginzburg-Landau theory; sound speed.

En este trabajo, luego de realizar una revisión de la teoría de las propiedades elásticas del compuesto  $\text{Sr}_2\text{RuO}_4$ , se presenta una extensión que permite explicar los resultados de los experimentos, sobre la rapidez del sonido realizados por Lupien y colaboradores [2] y Clifford y colaboradores [3]. Se muestra que la discontinuidad observada en la constante elástica  $C_{66}$  constituye una evidencia experimental directa de que el parámetro de orden  $\Psi$  tiene dos componentes y rompe la simetría de inversión temporal. También se realiza un estudio detallado del comportamiento elástico usando una teoría fenomenológica basada en el formalismo de Ginzburg-Landau.

**Descriptores:** Propiedades elásticas; superconductores no convencionales; simetría de inversión temporal; teoría de Ginzburg-Landau; rapidez del sonido.

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### 1. Introduction

In a triplet superconductor the electrons in the Cooper pairs are bound with spins parallel rather than antiparallel to one another, *i.e.* they are bound in spin triplets [5, 7, 13]. For this kind of superconductors, the spins are lying on the basal plane, while the pair orbital momentum is directed along the z-direction and their order parameter  $\Psi$  is represented by a three-dimensional vector  $\mathbf{d}(\mathbf{k})$ . If  $\Psi$  is of the type  $k_x \pm i k_y$ , there is a Cooper pair residual orbital magnetism, which gives place to an state of broken time reversal symmetry, edge currents in the surface of the superconductor, and a tiny magnetic field around non-magnetic impurities.

Based on the results of the Knight shift experiment performed through the superconducting transition temperature  $T_c$  [8, 9], it has been proposed that  $\text{Sr}_2\text{RuO}_4$  is a triplet superconductor. These experiments showed that Pauli spin susceptibility of the conduction electrons in the superconducting state remains unchanged respect to its value in the normal state. Moreover, it has been reported [10] that  $\Psi$  breaks time reversal symmetry, which constitutes another key feature of unconventionality.

The  $\text{Sr}_2\text{RuO}_4$  elastic constants  $C_{ij}$  have been measured as the temperature  $T$  is lowered through  $T_c$ . The results show a discontinuity in one of the elastic constants [2]. This im-

plies that  $\Psi$  has two different components with the time reversal symmetry broken. Similar conclusions from a muon spin relaxation ( $\mu\text{SR}$ ) experiment were reported by Luke *et al.* [10]. Recently, experiments on the effects of uniaxial stress  $\sigma_i$ , as a symmetry-breaking field were performed by Clifford and collaborators [3], reporting that for  $\text{Sr}_2\text{RuO}_4$  the symmetry-breaking field can be controlled experimentally. Additionally, experiments by Lupien *et al.* [2] showed the existence of small step in the transverse sound mode T[100].

This body of results evidences the need of extending or developing theoretical models to explain the changes occurring in  $C_{ij}$  at  $T_c$ , which, as far as we know, has not been carried out even in quite recent works [3]. Thus, the aim of our work is to extend an elasticity property phenomenological theory to show that  $\text{Sr}_2\text{RuO}_4$  is an unconventional superconductor with a two-component  $\Psi$  [4, 11]. Here, let us mention that a different theory of  $\text{Sr}_2\text{RuO}_4$  elastic properties was presented by Sigrist [12]. However, unlike this paper, Sigrist work does not take into account the splitting of  $T_c$  due to  $\sigma_i$ , and directly calculates the jumps at zero stress, where the derivative of  $T$  with respect to  $\sigma_i$  doesn't exist.

In this work, we first perform an analysis based on a  $\Psi$  that transforms as one of the two dimensional irreducible representations of the  $\text{Sr}_2\text{RuO}_4$  point group [4, 13]. Subsequently, we construct the  $\text{Sr}_2\text{RuO}_4$  superconducting phase

diagram under an external  $\sigma_i$ . This phase diagram is employed to develop a complete theory of the elastic behavior of  $\text{Sr}_2\text{RuO}_4$ , based on a two component Ginzburg-Landau (GL) model. This allows to properly calculate the jumps in the components of the elastic compliances  $S_{ij}$ . Finally, we propose that there are significant advantages for using  $\text{Sr}_2\text{RuO}_4$  as a material for a detailed study of symmetry-breaking effects in superconductivity described by a two-component  $\Psi$ .

## 2. Ehrenfest relations for a uniaxial stress $\sigma_i$

Provided that  $\sigma_i$  does not split the phase transition [4], for applied  $\sigma_i$ , Ehrenfest relations can be derived in analogous manner to the case of applied hydrostatic pressure [11, 14], under the condition that  $T_c$  is known as a function of  $\sigma_i$ . In order to simplify the calculations, we make use of the Voigt notation  $i = xx, yy, zz, yz, xz, xy$  [15].

For a second order phase transition, the Gibbs free energy  $G$  derivatives respect to  $T$ , the entropy  $S = -(\partial G/\partial T)_\sigma$ , and respect to  $\sigma_i$ , the elastic strain  $e_i = -(\partial G/\partial \sigma_i)_T$  are continuous functions of  $\sigma_i$  and  $T$ . Therefore, at the transition line,  $\Delta e_i(T, \sigma_j) = 0$  and  $\Delta S(T, \sigma_j) = 0$ . From this, for  $S$  and  $e_i$ , the boundary conditions between the two phases are

$$\begin{aligned} \Delta \left[ \left( \frac{\partial S}{\partial T} \right)_{\sigma_j} \right] dT + \Delta \left[ \left( \frac{\partial S}{\partial \sigma_j} \right)_T \right] d\sigma_j &= 0 \\ \Delta \left[ \left( \frac{\partial e_i}{\partial T} \right)_{\sigma_j} \right] dT + \Delta \left[ \left( \frac{\partial e_i}{\partial \sigma_j} \right)_T \right] d\sigma_j &= 0 \end{aligned} \quad (1)$$

By using the definitions of the thermal expansion  $\alpha_i = (\partial e_i/\partial T)_\sigma$ , the specific heat at constant stress,  $C_\sigma = T(\partial S/\partial T)_\sigma$ , and the elastic compliances  $S_{ij} = (\partial e_i/\partial \sigma_j)_T$ , together with the Maxwell identity  $(\partial S/\partial \sigma_i)_T = (\partial e_i/\partial T)_{\sigma_i}$ , the previous relations can be rewritten as,

$$\begin{aligned} \Delta \frac{C_\sigma}{T} + \frac{d\sigma_i}{dT} \Delta(\alpha_i)_{\sigma_i} &= 0 \\ \Delta(\alpha_i)_{\sigma_j} + \frac{d\sigma_j}{dT} \Delta(S_{ij})_T &= 0. \end{aligned} \quad (2)$$

From the first expression in Eq. (2), the relation for  $\alpha_i$  is found to be

$$\Delta \alpha_i = -\Delta C_\sigma \frac{d \ln T_c(\sigma_i)}{d\sigma_i}, \quad (3)$$

likewise, from the second expression of Eq. (2), the relation for  $S_{ij}$  is obtained to be,

$$\Delta S_{ij} = -\Delta \alpha_i \frac{dT_c(\sigma_j)}{d\sigma_j}. \quad (4)$$

It is important to distinguish that the print letter  $S$  denotes the entropy, while the symbol  $S_{ij}$  means the elastic compliances. In similar manner, the print letter  $C$  stands for the specific heat and the symbol  $C_{ij}$  for the elastic stiffness. Let us also point out that in deriving these expressions, we used

the fact that for a given thermodynamic quantity  $Q$ , its discontinuity along the transition line points is obtained from  $\Delta Q = Q(T_c + 0^+) - Q(T_c - 0^+)$ , where  $0^+$  is a positive infinitesimal quantity. Finally, by combining Eqs. (3) and (4), the variation in  $S_{ij}$  is found to be:

$$\Delta S_{ij} = \frac{\Delta C_\sigma}{T_c} \frac{dT_c(\sigma_i)}{d\sigma_i} \frac{dT_c(\sigma_j)}{d\sigma_j}. \quad (5)$$

Before continuing, it is interesting to mention that besides of our previous works [4, 11], we are not aware of any other works that have derived Ehrenfest relations for the case where applied  $\sigma_i$  produces a phase transition splitting.

## 3. Ginzburg-Landau model

In this section, a phenomenological model which takes into account the  $\text{Sr}_2\text{RuO}_4$  crystallographic point group  $D_{4h}$  is derived and employed. As we show, the analysis of  $G$ , using an order parameter which belongs to any of the one dimensional representations of  $D_{4h}$  is not able to describe the splitting of  $T_c$  under an external stress field. In order to account properly for the splitting, superconductivity in  $\text{Sr}_2\text{RuO}_4$  must be described by a  $\Psi$ , transforming as one of the  $D_{4h}$  two dimensional irreducible representations,  $E_{2g}$  or  $E_{2u}$ , which at this level of theoretical description render identical results [4, 11].

### 3.1. Superconducting free energy

In order to derive a suitable GL free energy  $G^\Gamma$ , we first will suppose that the  $\text{Sr}_2\text{RuO}_4$  superconductivity is described by an order parameter  $\psi^\Gamma$ , which transforms according to one of the eight one-dimensional representations of  $D_{4h}$ :  $\Gamma = A_{1g}, A_{2g}, B_{1g}, B_{2g}, A_{1u}, A_{2u}, B_{1u},$  or  $B_{2u}$ . Let us notice that an analysis employing the  $D_4$  point group renders similar results. Here we will analyze the terms in  $G^\Gamma$  linear in  $\sigma_i$  and quadratic in  $\psi^\Gamma$ :

$$\begin{aligned} G^\Gamma &= G_0 + \alpha(T) |\psi^\Gamma|^2 + \frac{b}{2} |\psi^\Gamma|^4 \\ &+ [a(\sigma_{xx} + \sigma_{yy}) + c\sigma_{zz}] |\psi^\Gamma|^2. \end{aligned} \quad (6)$$

The terms proportional to  $\sigma_{xx}$ ,  $\sigma_{yy}$  and  $\sigma_{zz}$  in Eq. (6) give rise to discontinuities in the elastic constants, evidenced from sound speed measurements [17]. On the other hand, discontinuities in the elastic compliance  $S_{66}$  and in the elastic constant  $C_{66}^i$  arise from the linear coupling with  $\sigma_{xy}$ . However, due to symmetry, the later linear coupling does not exist for any  $\Gamma$ ; therefore,  $S_{66}$  and  $C_{66}$  are expected to be continuous at  $T_c$  for any of the one-dimensional irreducible representation that assumes a one-dimensional  $\psi^\Gamma$ . Nevertheless, the results of Lupien *et al.* experiments [2] showed a discontinuity in  $C_{66}$ . Hence, based exclusively on sound speed measurements, we conclude that none of the one-dimensional irreducible representations can provide an appropriate description of superconductivity in  $\text{Sr}_2\text{RuO}_4$ . As far as we know, this conclusion has not been previously established in the literature [3]. Let us mention that for any one-dimensional  $\Gamma$ ,

a detailed analysis of the calculation of the jumps in  $C_{66}$  is presented in Ref. 11.

Due to the absence of discontinuity in  $S_{66}$  for any of the one-dimensional  $\Gamma$ , the superconductivity in  $\text{Sr}_2\text{RuO}_4$  must be described by an order parameter  $\psi^E$  transforming as one of the two-dimensional representations  $E_{2g}$  or  $E_{2u}$  [4]. The  $GL$  theory establishes that only the parameters of one of the irreducible representations becomes non-zero at  $T_c$ . Therefore, following the evidence provided in Refs. 5 and 19, we choose the  $E_{2u}$  spin-triplet state as the correct representation for  $\text{Sr}_2\text{RuO}_4$ , and the speed measurements are analyzed in terms of the model  $\psi^E = (\psi_x, \psi_y)$ , with  $\psi_x$  and  $\psi_y$  transforming as the components of a vector in the basal plane. The expression for  $G$  is determined by symmetry arguments based on the analysis of the second and fourth order invariants (real terms) of  $G^\Gamma$ . To maintain gauge symmetry, only real and even products of  $\Psi$  can occur in the expansion of  $G^\Gamma$ ; thus, we find that all real invariants should be formed by second and fourth order products of  $\psi^i$ . To obtain its expression, we use the fact that  $G$  is invariant with respect to a transformation by the generators  $c_{4z}$  and  $c_{2x}$  of  $D_{4h}$ . Applying the generators to different second and fourth order combination of products of  $\psi$ 's, we find only one second order invariant  $|\psi_x|^2 + |\psi_y|^2$  and three fourth order invariants, namely  $|\psi_x|^2|\psi_y|^2$ ,  $|\psi_x|^4 + |\psi_y|^4$ , and  $\psi_x^2\psi_y^{*2} + \psi_x^{*2}\psi_y^2$ .

For the zero  $\sigma_i$  case, the expansion of  $G$  gives place to:

$$G = G_0 + \alpha(T) (|\psi_x|^2 + |\psi_y|^2) + \frac{b_1}{4} (|\psi_x|^2 + |\psi_y|^2)^2 + b_2|\psi_x|^2|\psi_y|^2 + \frac{b_3}{2} (\psi_x^2\psi_y^{*2} + \psi_y^2\psi_x^{*2}), \quad (7)$$

where  $\alpha = \alpha'(T - T_{c0})$  and the coefficients  $b_1$ ,  $b_2$ , and  $b_3$  are material-dependent real constants [20, 21]. These coefficients have to satisfy special conditions in order to maintain the free energy stability. The analysis of  $G$  is accomplished by considering two component  $(\psi_x, \psi_y)$  with the form:

$$(\psi_x, \psi_y) = (\eta_x e^{i\varphi/2}, i \eta_y e^{-i\varphi/2}); \quad (8)$$

where  $\eta_x$  and  $\eta_y$  are both real and larger than zero. After substitution of  $\psi_x$  and  $\psi_y$  in Eq. (7),  $G$  becomes:

$$G = G_0 + \alpha(T)(\eta_x^2 + \eta_y^2) + \frac{b_1}{4}(\eta_x^2 + \eta_y^2)^2 + (b_2 - b_3)\eta_x^2\eta_y^2 + 2b_3\eta_x^2\eta_y^2 \sin^2 \varphi. \quad (9)$$

For fixed values of the coefficients  $b_1$  and  $b_2$ , if  $b_3 > 0$ ,  $G$  will reach a minimal value if the last term vanishes, *i.e.* if  $\varphi = 0$ . Moreover, if  $\eta_x$  and  $\eta_y$  have the form  $\eta_x = \eta \sin \chi$  and  $\eta_y = \eta \cos \chi$ ,  $G$  becomes

$$G = G_0 + \alpha(T)\eta^2 + \frac{b_1}{4}\eta^4 - \frac{\tilde{b}}{4}\eta^4 \sin^2 2\chi, \quad (10)$$

where  $\tilde{b} \equiv b_3 - b_2$ . If  $\tilde{b} > 0$ ,  $G$  reaches its minimum value if  $\sin^2 2\chi = 1$ , this condition is satisfied if  $\chi = \pi/4$ ; and therefore  $\eta_x = \eta_y$ . On the other hand, if  $\tilde{b} < 0$ , then  $G$  becomes

minimal if  $\sin^2 2\chi = 0$ . In this case, either  $\eta_x = 0$  or  $\eta_y = 0$ . Since for a superconducting state  $(\psi_x, \psi_y) \sim (1, \pm i)$ , from the previous analysis, the lowest  $G$  state corresponds to  $b_3 - b_2 > 0$ . This thermodynamic state breaks time-reversal-symmetry; and hence, it is believed to be the state describing superconductivity in  $\text{Sr}_2\text{RuO}_4$  [4, 5, 7]. In addition, it is found that for the phase transition to be of second order, it is required that  $b \equiv b_1 + b_2 - b_3 > 0$ .

At this point it is important to understand why the state  $(\psi_x, \psi_y) \sim (1, \pm i\epsilon)$  has been chosen for the analysis of  $\sigma_6$  and why it gives rise to the discontinuity in  $S_{66}$  [11]. Minimization of Eq. (7) with respect to  $\varphi$  and  $\chi$ , and employing Eq. (8) renders a set of solutions for the two-component order parameter which depend on the relation between the coefficients  $b_1$ ,  $b_2$ , and  $b_3$  and also on the value of the phases  $\varphi$  and  $\chi$ . Thus, for the E representation, solutions of the form,

$$\psi_1 = \eta (1, 0) e^{i\varphi}, \quad (11)$$

are obtained, which are very similar to those found for the  $D_4$  one-dimensional irreducible representation. Therefore, these solutions are not able to account for the jump in  $C_{66}$ . However, solutions with both components different than zero are also attained:

$$\psi_2 = \frac{\sqrt{2}}{2} e^{i\pi/4} (1, 1) \eta, \quad \psi_3 = \frac{\sqrt{3}}{2} (1, i) \eta. \quad (12)$$

Each of these solutions corresponds to different relations for the  $b_i$ . This is illustrated by Fig. 1, which shows the phase diagram, displaying the domains of  $\psi_1$ ,  $\psi_2$  and  $\psi_3$  as a function of  $b_1$ ,  $b_2$  and  $b_3$ . Now, if the jump in  $C_{66}$  corresponds to a  $G$  minimum, the coupling term with  $\sigma_6$  must be taken to be different from zero. If the solution  $\psi_2$  is considered, the term containing  $\sigma_6$  becomes zero; therefore it is not acceptable. On the other hand, this requirement is satisfied by  $\Psi_3$ , with the form  $(1, i)\eta$ . Hence, the  $GL$  analysis renders  $\Psi_3$  as the solution that breaks time reversal symmetry.

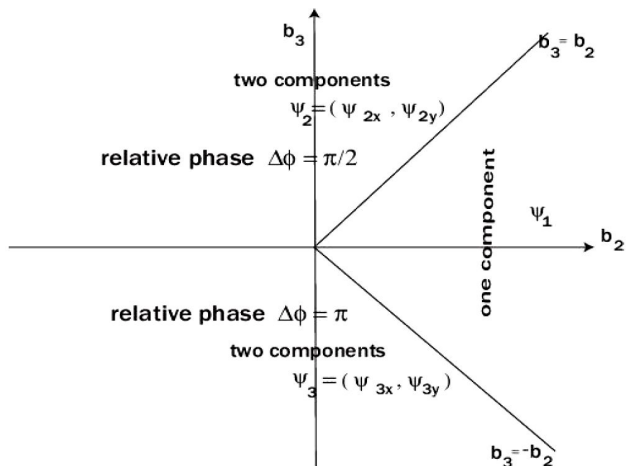


FIGURE 1. Superconducting state phase diagram for the two dimensional representation E of the tetragonal group  $D_4$  as function of the material parameters  $b_2$  and  $b_3$  showing the domains which correspond to the order parameters  $\psi_1$ ,  $\psi_2$ , and  $\psi_3$ . Each domain corresponds to a different superconducting class.

### 3.2. Coupling of the order parameter to an external stress

The transition to an unconventional superconducting state shows manifestations as the breakdown of symmetries, such as the crystal point group or the time reversal symmetry [20,21]. This loss of symmetry has measurable manifestations in observable phenomena, as the splitting of  $T_c$  under an elastic deformation. The coupling between the crystal lattice and the superconducting state is described Refs. 20 and 21. As explained there, close to  $T_c$ , a new term is added to  $G$ , which couples in second order  $\Psi$  with  $e_{ij}$  and in first order  $\Psi$  with  $\sigma_{ij}$ . These couplings give place to discontinuities in  $S_{ijkl}$  at  $T_c$ .

### 3.3. Analysis of the phase diagram

An expression for  $G$  accounting for a phenomenological coupling to  $C_{66}$  in the  $\text{Sr}_2\text{RuO}_4$  basal plane is given by

$$G = G_0 + \alpha'(T - T_{c0})(|\psi_x|^2 + |\psi_y|^2) + b_2|\psi_x|^2|\psi_y|^2 + \frac{b_1}{4}(|\psi_x|^2 + |\psi_y|^2)^2 + \frac{b_3}{2}(\psi_x^2\psi_y^{*2} + \psi_y^2\psi_x^{*2}) - \frac{1}{2}S_{ij}\sigma_i\sigma_j + \sigma_i\Lambda_i + \sigma_id_{ij}E_j. \quad (13)$$

Here,  $\Lambda_i$  are the temperature-dependent  $\alpha_i$ ,  $d_{ij}$  are the coupling terms between  $\Psi$  and  $S_{ij}$  and  $E_j$  are the invariant elastic compliance tensor components, defined below. In order to determine these invariants describing the coupling of the order parameter to the stress tensor, we construct the tensor  $E_j$  with Voigt components  $E_1 = |\psi_x|^2$ ,  $E_2 = |\psi_y|^2$  and  $E_6 = \psi_x^*\psi_y + \psi_x\psi_y^*$ ; where  $E_6$  couples  $\sigma_6$  and  $\Psi$ . The tensor  $d_{ij}$  couples  $E_i$  with  $\sigma_j$  and has the same nonzero components as  $S_{ij}$ . By applying symmetry considerations [4], it is shown that the only non-vanishing independent components of  $d_{ij}$  are  $d_{11}$ ,  $d_{12} = d_{21}$ ,  $d_{31} = d_{32}$ , and  $d_{66}$ . Contributions to  $G$  that are quadratic in both,  $\Psi$  and  $\sigma_6$  were neglected. Such terms would have given an additional  $T$  dependence to the  $S_{ij}$  [17]. However, given the large number of independent constants occurring in the associated sixth rank tensor, at this point, it is not clear whether or not the explicit inclusion of such terms would be productive.

Now, let us consider the case of uniaxial compression along the  $a$  axis (only with  $\sigma_1 < 0$ ). If in Eq. (13), only quadratic terms in  $\Psi$  are kept, this equation can be written as

$$G_{\text{quad}} = \alpha'[T - T_{c+}(\sigma_1)]|\psi_x|^2 + \alpha'[T - T_{cy}(\sigma_1)]|\psi_y|^2, \quad (14)$$

here  $T_{c+}(\sigma_1)$  and  $T_{cy}(\sigma_1)$  are given by

$$T_{c+}(\sigma_1) = T_{c0} - \frac{\sigma_1 d_{11}}{\alpha'}, \quad T_{cy}(\sigma_1) = T_{c0} - \sigma_1 \frac{d_{12}}{\alpha'}. \quad (15)$$

In what follows, we assume that  $d_{11} - d_{12} > 0$ , such that  $T_{c+} > T_{cy}$ . Notice that this does not imply any loss in generality, assuming  $d_{11} - d_{12} < 0$ , would render an identical model, simply by exchanging the  $x$  and  $y$  indices. Here,  $T_{c+}$

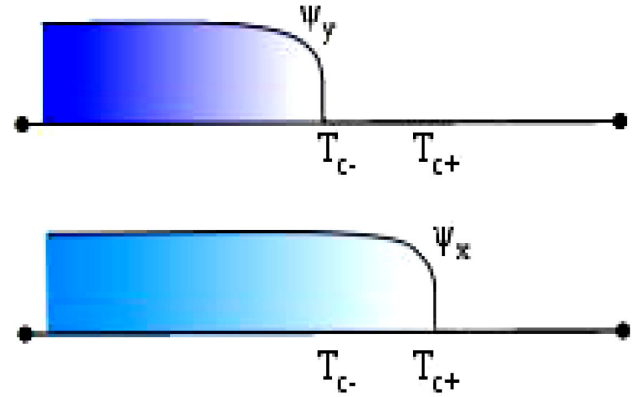


FIGURE 2. Temperature behavior of the two component order parameter  $(\psi_x, \psi_y)$  for the case of a nonzero uniaxial stress below  $T_c$ . Notice that only the BCS component  $\psi_x(T_{c+})$  becomes non zero for temperatures between  $T_{c+}$  and  $T_{c-}$ . The second unconventional component  $\psi_y(T_{c-})$  only appears below  $T_{c-}$ .

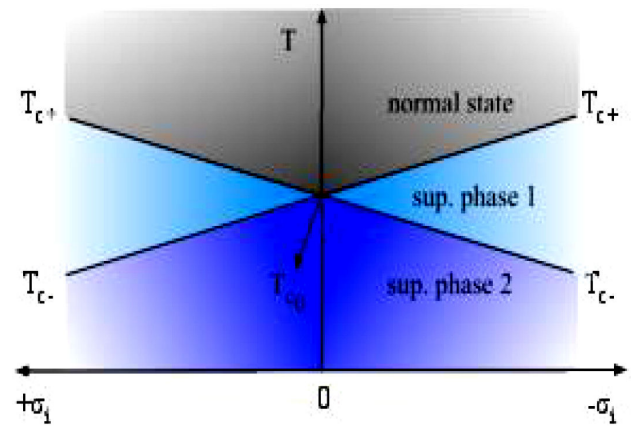


FIGURE 3. Phase diagram showing the upper and lower superconducting transition temperatures,  $T_{c+}$  and  $T_{c-}$ , respectively, as functions of the compressible stress  $-\sigma_1$  along the  $a$  axis.

is the higher of the two critical temperatures at which the initial transition occurs. As should be expected, just below  $T_{c+}$ , only  $\psi_x$  is non zero. As  $T$  is further lowered, another phase transition happens at  $T_{c-}$ , which is different than  $T_{cy}$ . Below  $T_{c-}$ , the  $\psi_y$  is also different from zero (see Fig. (2)). Thus, in the presence of a non zero compressible  $\sigma_1$ ,  $\Psi$  has the form  $(\psi_x, \psi_y) \approx \psi(1, \pm i \epsilon)$ , where  $\epsilon$  is real and equal to zero between  $T_{c+}$  and  $T_{c-}$  (phase 1), and increases from  $\epsilon = 0$  to  $\epsilon \approx 1$  as  $T$  becomes smaller than  $T_{c-}$  (phase 2), as illustrated in Figs. (1) and (2).

The next step is finding  $T_{c-}$ . To achieve this goal, the equilibrium value of the non zero component of  $\psi_x$ ,  $\psi_x^2 = -2\alpha_x/b_1$  is replaced in Eq. (13) and  $T_{c-}$  follows from

$$T_{c+} - T_{c-} = - \left[ \frac{d_{11} - d_{12}}{2\alpha'} \right] \left[ \frac{\tilde{b} + b}{\tilde{b}} \right] \sigma_1. \quad (16)$$

To obtain Eq. (16), it is assumed that  $\sigma_1^2 \ll \sigma_1$  and only linear terms in  $\sigma_1$  are kept. The phase diagram for this system is shown in Fig. (3).

### 4. Calculation of the discontinuities

As discussed before, an external uniaxial stress acting on the Sr<sub>2</sub>RuO<sub>4</sub> basal plane breaks the tetragonal symmetry of the crystal. As a consequence of this, when a second order transition to the superconducting state occurs, it splits into two transitions. For the case of applied σ<sub>1</sub>, the analysis of the behavior of the sound speed at T<sub>c</sub> requires a systematic study of these second-order phase transitions. Moreover, thermodynamic quantities, such as dT<sub>c</sub>/dσ<sub>i</sub>, C<sub>σ</sub>, and α<sub>σ</sub>, which are needed in order to calculate the components S<sub>ij</sub><sup>σ</sup>, are accompanied by a discontinuity at each of the second order phase transitions.

As depicted in Fig. (3), for a given σ<sub>1</sub> ≠ 0 as T is lowered below T<sub>c+</sub>, a first discontinuity for a thermodynamical quantity Q is observed at the first line of transition temperatures. This discontinuity along the transition line, corresponding to the higher transition temperatures, T = T<sub>c+</sub> (σ<sub>i</sub>) is given by ΔQ<sup>+</sup> = Q(T<sub>c+</sub> + 0<sup>+</sup>) - Q(T<sub>c+</sub> - 0<sup>+</sup>), where 0<sup>+</sup> is a positive infinitesimal number. If T is further dropped below T<sub>c-</sub>, a second discontinuity arises, and the lower line of transition temperatures appears. The discontinuity along this line, at T = T<sub>c-</sub> (σ<sub>i</sub>), is defined by ΔQ<sup>-</sup> = Q(T<sub>c-</sub> + 0<sup>+</sup>) - Q(T<sub>c-</sub> - 0<sup>+</sup>) [18]. The sum of these two discontinuities

$$\Delta Q(T_{c0}, \sigma = 0) = \Delta Q^+ + \Delta Q^-, \tag{17}$$

gives the correct expressions for the discontinuities at T<sub>c0</sub>, for the case with σ<sub>i</sub>=0, where the Ehrenfest relations do not hold directly [4]. As an example of these discontinuities, the two jumps in C<sub>σ</sub> under an external σ<sub>i</sub> are sketched in Fig. (4).

#### 4.1. Jumps due to a uniaxial stress σ<sub>1</sub>

The free energy, Eq. (13), for the cases where both σ<sub>1</sub> and σ<sub>6</sub> are nonzero is:

$$\begin{aligned} G = & G_0 + \alpha_x |\psi_x|^2 + \alpha_y |\psi_y|^2 + \sigma_6 d_{66} (\psi_x \psi_y^* + \psi_x^* \psi_y) \\ & + \frac{b_1}{4} (|\psi_x|^2 + |\psi_y|^2)^2 + b_2 |\psi_x|^2 |\psi_y|^2 \\ & + \frac{b_3}{2} (\psi_x^2 \psi_y^{*2} + \psi_y^2 \psi_x^{*2}). \end{aligned} \tag{18}$$

Here α<sub>x</sub> = α'(T - T<sub>c0</sub>) + σ<sub>1</sub>d<sub>11</sub> and α<sub>y</sub> = α'(T - T<sub>c0</sub>) + σ<sub>1</sub>d<sub>12</sub>. If only σ<sub>1</sub> is applied, this equation becomes:

$$\begin{aligned} \Delta G = & \alpha_x |\psi_x|^2 + \alpha_y |\psi_y|^2 + \frac{b_1}{4} (|\psi_x|^2 + |\psi_y|^2)^2 \\ & + b_2 |\psi_x|^2 |\psi_y|^2 + \frac{b_3}{2} (\psi_x^2 \psi_y^{*2} + \psi_y^2 \psi_x^{*2}), \end{aligned} \tag{19}$$

where ΔG = G - G<sub>0</sub>(T). The nature of the superconducting state that follows from Eq. (19), depends on the values of the coefficients b<sub>1</sub>, b<sub>2</sub>, and b<sub>3</sub>. The analysis from Eq. (19) of the superconducting part of G is performed by using, as was done previously, an expression for Ψ given by Eq. (8).

At T<sub>c+</sub> and in the presence of σ<sub>1</sub>, the second order terms in Eq. (19) dominate and Ψ has a single component ψ<sub>x</sub>; whereas at T<sub>c-</sub> a second component ψ<sub>y</sub> appears. Thus, at very low T, the fourth order terms dominate the Eq. (19) behavior. Each of these two-component domains has the form of ψ<sub>2</sub> given by Eq. (12). In this case, G can be written in terms of η<sub>x</sub> and η<sub>y</sub> as

$$\begin{aligned} \Delta G = & \alpha_x \eta_x^2 + \alpha_y \eta_y^2 + \frac{b_1}{4} (\eta_x^2 + \eta_y^2)^2 \\ & + (b_2 - b_3) \eta_x^2 \eta_y^2 + 2b_3 \eta_x^2 \eta_y^2 \sin^2 \varphi. \end{aligned} \tag{20}$$

The analysis of Eq. (20) depends on the relation between the coefficients b<sub>1</sub>, b<sub>2</sub>, and b<sub>3</sub>. Assuming that b<sub>3</sub> > 0, and η<sub>x</sub> and η<sub>y</sub> are both different from zero, and following the procedure described after Eq. (9) one arrives to

$$(\psi_x, \psi_y) \approx (1, \pm i\epsilon), \tag{21}$$

where ε is real and grows from ε = 0 to ε ≈ 1 as T is reduced below T<sub>c-</sub>, while Eq. (20) becomes

$$\Delta G = \alpha_x \eta_x^2 + \alpha_y \eta_y^2 + \frac{b_1}{4} (\eta_x^2 + \eta_y^2)^2 - (b_3 - b_2) \eta_x^2 \eta_y^2. \tag{22}$$

To calculate the jumps at T<sub>c+</sub>, we use α<sub>x</sub> = α'(T - T<sub>c+</sub>) and α<sub>y</sub> = α'(T - T<sub>cy</sub>), and assume that T<sub>c+</sub> > T<sub>cy</sub>. For the interval T<sub>c+</sub> > T > T<sub>c-</sub>, the equilibrium value for Ψ satisfies α<sub>x</sub> > 0 and α<sub>y</sub> = 0, i.e. η<sub>x</sub> > 0 and η<sub>y</sub> = 0, with η<sub>x</sub><sup>2</sup> = -2α<sub>x</sub>/b<sub>1</sub>, obtaining that T<sub>c+</sub> and its derivative with respect to σ<sub>1</sub> are respectively,

$$\begin{aligned} T_{c+}(\sigma_1) = & T_{c0} - \frac{\sigma_1}{\alpha'} d_{11}, \\ \frac{dT_{c+}}{d\sigma_1} = & - \frac{d_{11}}{\alpha'}. \end{aligned} \tag{23}$$

The specific heat discontinuity at T<sub>c+</sub>, relative to its normal state value, is calculated by using:

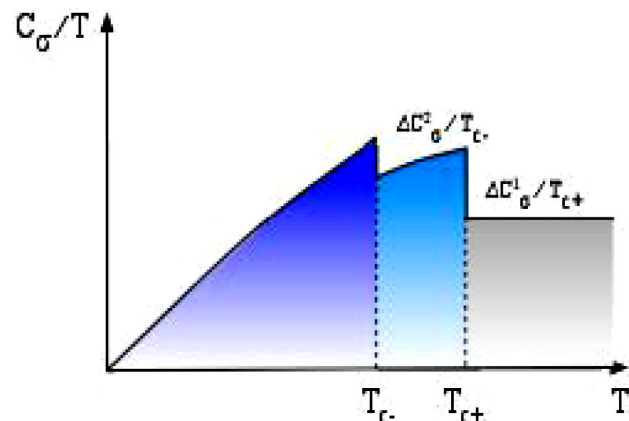


FIGURE 4. Schematic dependence of the specific heat on the temperature, for the case of an uniaxial stress splitting the Sr<sub>2</sub>RuO<sub>4</sub> transition Temperature. Notice the two jumps in the heat capacity near the transition temperatures T<sub>c+</sub> and T<sub>c-</sub>.

$$\Delta C_{\sigma_1} = -T \frac{\partial^2 \Delta G}{\partial^2 T} \Big|_{T=T_{c+}}, \quad (24)$$

and renders the result

$$\Delta C_{\sigma_1}^+ = -\frac{2 T_{c+} \alpha'^2}{b_1}. \quad (25)$$

A schematic depiction of the  $C_\sigma$  discontinuities below this transition temperature is exhibited in Fig. (4). At  $T_{c+}$ , the discontinuity in  $\alpha_\sigma$  is calculated by applying the Ehrenfest relation of Eq. (3), yielding:

$$\Delta \alpha_1^+ = -\frac{2\alpha' d_{11}}{b_1}. \quad (26)$$

The discontinuities in  $S_{ij}$  are obtained by using Eqs. (4) and (5), rendering the result,

$$\Delta S_{i'j'}^+ = -\frac{2 d_{i'1} d_{j'1}}{b_1}. \quad (27)$$

In the previous expression a prime on an index (as in  $i'$  or  $j'$ ) indicates a Voigt index taking only the values 1,2, or 3. Thus, from Eq. (27) the change in  $S_{11}$  at  $T_{c+}$  can be calculated to be  $\Delta S_{11}^+ = -\frac{2 d_{11}^2}{b_1}$ .

To find the discontinuities at  $T_{c-}$ , the invariant  $(\eta_x^2 + \eta_y^2)^2$  in Eq. (22) is expanded, after which  $G$  takes the form,

$$\Delta G = \alpha_x \eta_x^2 + \frac{b_1}{4} \eta_x^4 + \left[ \alpha_y + \left( \frac{b_1}{2} + b_2 - b_3 \right) \eta_x^2 \right] \eta_y^2 + \frac{b_1}{4} \eta_y^4. \quad (28)$$

In this expression, the second order term in  $\eta_y$  is renormalized by the square of  $\eta_x$ . The second transition temperature is determined from the zero of the total prefactor of  $\eta_y^2$ , obtaining that  $T_{c-}$  and its derivative with respect to  $\sigma_1$  are:

$$T_{c-}(\sigma_1) = T_c - \frac{\sigma_1}{2\alpha'} \left[ d_{11} + d_{12} - \frac{b}{\tilde{b}}(d_{12} - d_{11}) \right],$$

$$\frac{d T_{c-}}{d \sigma_1} = -\frac{1}{2\alpha'} \left[ d_{11} + d_{12} - \frac{b}{\tilde{b}}(d_{12} - d_{11}) \right]. \quad (29)$$

Below  $T_{c-}$  the superconducting free energy, Eq. (28) has to be minimized respect to both components of  $\Psi$ . After doing so,  $\eta_x$  and  $\eta_y$  for this temperature range are found to be

$$\eta_x^2 = -\frac{1}{2 b \tilde{b}} \left[ (b - \tilde{b})\alpha_y + (b + \tilde{b})\alpha_x \right],$$

$$\eta_y^2 = -\frac{1}{2 b \tilde{b}} \left[ (b - \tilde{b})\alpha_x + (b + \tilde{b})\alpha_y \right]. \quad (30)$$

This analysis shows that the second superconducting phase is different in symmetry, and that time reversal symmetry is broken. The change in  $C_{\sigma_1}$  at  $T_{c-}$ , with respect to its value in the normal phase,  $\Delta C_{\sigma_1}^{-,N}$ , is found to be,  $\Delta C_{\sigma_1}^{-,N} = -2 T_{c-} \alpha' 1/b$ . The specific heat variation at  $T_{c-}$  is,

$$\Delta C_{\sigma_1}^- = \Delta C_{\sigma_1}^{-,N} - \Delta C_{\sigma_1}^+, \quad (31)$$

which results in

$$\Delta C_{\sigma_1}^- = -2 T_{c-} \alpha'^2 \frac{\tilde{b}}{b b_1}. \quad (32)$$

The size of these jumps is complicated to infer, because it depends on the material parameters  $b_1, b_2$  and  $b_3$ , and on the coupling constants  $d_{11}$  and  $d_{12}$ .

With the help of the Ehrenfest relation, Eq. (3), the discontinuity in  $\alpha_i$  at  $T_{c-}$  is obtained to be

$$\Delta \alpha_{i'}^- = -\alpha' \frac{\tilde{b}}{b b_1} \left( d_{i'+} - \frac{b}{\tilde{b}} d_{i'-} \right), \quad (33)$$

and after employing Eqs. (4) and (5), the discontinuity in  $S_{i'j'}$  at  $T_{c-}$  is shown to be

$$\Delta S_{i'j'}^- = -\frac{\tilde{b}}{2bb_1} \left( d_{i'+} - \frac{b}{\tilde{b}} d_{i'-} \right) \left( d_{j'+} - \frac{b}{\tilde{b}} d_{j'-} \right). \quad (34)$$

Here  $d_{i'\pm} = d_{i'1} \pm d_{i'2}$ . The discontinuities occurring at  $T_{c0}$ , in the absence of uniaxial stress, can be obtained by adding the discontinuities occurring at  $T_{c+}$  and  $T_{c-}$ , yielding:

$$\Delta C_{\sigma_1}^0 = -\frac{2T_{c0}\alpha'^2}{b}, \quad \Delta \alpha_{i'}^0 = -\frac{\alpha' d_{i'+}}{b},$$

$$\Delta S_{i'j'}^0 = -\frac{1}{2} \left( \frac{d_{i'+} d_{j'+}}{b} + \frac{d_{i'-} d_{j'-}}{\tilde{b}} \right). \quad (35)$$

Before continuing, it is important to emphasize that at zero stress, the derivative of  $T_c$  with respect to  $\sigma_i$  is not defined; therefore, there is no reason to expect any of the Ehrenfest relations to hold [4,11].

#### 4.2. Jumps due to a shear stress $\sigma_6$

When a shear stress  $\sigma_6$  is applied to the basal plane of  $\text{Sr}_2\text{RuO}_4$ , the crystal tetragonal symmetry is broken, and a second transition to a superconducting state occurs. Accordingly, for this case the analysis of the sound speed behavior at  $T_c$  also requires a systematic study of the two successive second order phase transitions. Very important to mention that the  $C_{66}$  discontinuity observed by Lupien [2] at  $T_c$ , can be explained in this context.

If there is a double transition, the derivative of  $T_c$  with respect to  $\sigma_6$  *i.e.*  $dT_c/d\sigma_6$  is different for each of the two transition lines. At each of these transitions,  $C_{\sigma_6}$ ,  $\alpha_{\sigma_6}$ , and  $S_{ij}^{\sigma_6}$  show discontinuities. As discussed before, the sum of them gives the correct expressions for the discontinuities at zero shear stress, where the Ehrenfest relations do not hold.

The  $T_c - \sigma_6$  phase diagram will be similar to that obtained for  $\sigma_1$ ; therefore, the diagram in Fig. (3) also qualitatively holds here. In the case of an applied  $\sigma_6$ ,  $\Delta G$  is given by

$$\Delta G = \alpha (|\psi_x|^2 + |\psi_y|^2) + \sigma_6 d_{66} (\psi_x \psi_y^* + \psi_x^* \psi_y)$$

$$+ \frac{b_1}{4} (|\psi_x|^2 + |\psi_y|^2)^2 + b_2 |\psi_x|^2 |\psi_y|^2$$

$$+ \frac{b_3}{2} (\psi_x^2 \psi_y^{*2} + \psi_y^2 \psi_x^{*2}). \quad (36)$$

Here  $\alpha = \alpha'(T - T_{c0})$ , and the minimization of  $\Delta G$  is performed as in the  $\sigma_1$  case, *i.e.* by substituting the general expression for  $\Psi$  given in Eq. (8). After doing so,  $\Delta G$  becomes

$$\Delta G = \alpha(\eta_x^2 + \eta_y^2) + 2\eta_x\eta_y \sigma_6 \sin \varphi d_{16} + \frac{b_1}{4} (\eta_x^2 + \eta_y^2)^2 + (b_2 - b_3)\eta_x^2\eta_y^2 + 2b_3\eta_x^2\eta_y^2 \sin^2 \varphi. \quad (37)$$

In the presence of  $\sigma_6$ , the second order term determines the phase below  $T_{c+}$ , which is characterized by  $\psi_x$  and by  $\psi_y = 0$ . As the temperature is lowered below  $T_{c-}$ , depending of the value of  $b_3$  a second component  $\psi_y$  may appear. If at  $T_{c-}$  a second component occurs, the fourth order terms in Eq. (37) will be the dominant one. Thus for very low  $T$ 's, or for  $\sigma_6 \rightarrow 0$ , a time-reversal symmetry-breaking superconducting state emerges. The analysis of Eq. (37) depends on the relation between the coefficients  $b_2$  and  $b_3$ . It also depends on the values of the quantities  $\eta_x$  and  $\eta_y$ , and of the phase  $\varphi$ . If  $b_3 < 0$ , and  $\eta_x$  and  $\eta_y$  are both nonzero, the state with minimum energy has a phase  $\varphi = \pi/2$ . The transition temperature is obtained from Eq. (37), by performing the canonical transformations:  $\eta_x = (1/\sqrt{2})(\eta_\mu + \eta_\xi)$  and  $\eta_y = (1/\sqrt{2})(\eta_\mu - \eta_\xi)$ . After their substitution, Eq. (37) becomes

$$\Delta G = \alpha_+\eta_\xi^2 + \alpha_-\eta_\mu^2 + \frac{1}{4}(\eta_\xi^2 + \eta_\mu^2)^2 + (b_2 + b_3)(\eta_\xi^2 - \eta_\mu^2)^2. \quad (38)$$

If, as was done before,  $\eta_\xi = \eta \sin \chi$  and  $\eta_\mu = \eta \cos \chi$ , Eq. (38) takes the form

$$\Delta G = \alpha_+\eta^2 \sin^2 \chi + \alpha_-\eta^2 \cos^2 \chi + \frac{\eta^4}{4} [b_1 + (b_2 + b_3) \cos^2 2\chi]. \quad (39)$$

$\Delta G$  is minimized if  $\cos 2\chi = 1$ , this is, if  $\chi = 0$ . Also, in order for the phase transition to be of second order,  $b'$ , defined as  $b' \equiv b_1 + b_2 + b_3$ , must be larger than zero. Therefore, if  $\sigma_6$  is non zero, the state with the lowest free energy corresponds to  $b_3 < 0$ , phase  $\varphi$  equal to  $\pi/2$ , and  $\Psi$  of the form:

$$(\psi_x, \psi_y) \approx \eta (e^{i\frac{\varphi}{2}}, e^{-i\frac{\varphi}{2}}). \quad (40)$$

In phase 1 of Fig. (3),  $\varphi = 0$ , and as  $T$  is lowered below  $T_{c-}$ , phase 2,  $\varphi$  grows from 0 to approximately  $\pi/2$ . Again, following an analysis similar to that carried out for  $\sigma_1$ , the two transition temperatures  $T_{c+}$  and  $T_{c-}$  are obtained to be:

$$T_{c+}(\sigma_6) = T_{c0} - \frac{\sigma_6}{\alpha'} d_{66},$$

$$T_{c-}(\sigma_6) = T_{c0} + \frac{b}{2b_3 \alpha'} \sigma_6 d_{66}. \quad (41)$$

The derivative of  $T_{c+}$  with respect to  $\sigma_6$ , and the discontinu-

ity in  $C_{\sigma_6}^+$  at  $T_{c+}$  are respectively found to be:

$$\frac{dT_{c+}}{d\sigma_6} = -\frac{d_{66}}{\alpha'},$$

$$\Delta C_{\sigma_6}^+ = -\frac{2T_{c+} \alpha'^2}{b'}. \quad (42)$$

After applying the Ehrenfest relations, Eqs. (4) and (5), the results for  $\Delta\alpha_{\sigma_6}$  and  $\Delta S_{66}$  at  $T_{c+}$  are:

$$\Delta\alpha_{\sigma_6}^+ = -\frac{2\alpha' d_{66}}{b'},$$

$$\Delta S_{66}^+ = -\frac{2d_{66}^2}{b'}. \quad (43)$$

For  $T_{c-}$ , the derivative of this transition temperature with respect to  $\sigma_6$ , and the discontinuities in the specific heat, thermal expansion and elastic stiffness respectively are:

$$\frac{dT_{c-}}{d\sigma_6} = \frac{b d_{66}}{2b_3 \alpha'},$$

$$\Delta C_{\sigma_6}^- = -\frac{4T_{c-} \alpha'^2 b_3}{b b'}, \quad (44)$$

$$\Delta\alpha_{\sigma_6}^- = \frac{2\alpha' d_{66}}{b'},$$

$$\Delta S_{66}^- = -\frac{d_{66}^2 b}{b' b_3}. \quad (45)$$

Since for the case of  $\sigma_6$ , the derivative of  $T_c$  with respect to  $\sigma_6$  is not defined at zero stress point, the Ehrenfest relations do not hold at  $T_{c0}$ . Thus, the discontinuities occurring at  $T_{c0}$ , in the absence of  $\sigma_6$ , are calculated by adding the expressions obtained for the discontinuities at  $T_{c+}$  and  $T_{c-}$ ,

$$\Delta C_{\sigma_6}^0 = -\frac{2T_{c0} \alpha'^2}{b},$$

$$\Delta S_{66}^0 = -\frac{d_{66}^2}{b_3}, \quad (46)$$

$$\Delta\alpha_{\sigma_6}^0 = 0.$$

Notice that in this case, there is no discontinuity for  $\alpha_{\sigma_6}^0$ .

Since the phase diagram was determined as a function of  $\sigma_6$ , rather than as a function of the strain, (see Fig. (3)), in this work, as in Refs. 4 and 11, we make use of the  $6 \times 6$  elastic compliance matrix  $S$ , whose matrix elements are  $S_{ij}$ . However, the sound speed measurements are best interpreted in terms of the elastic stiffness matrix  $C$ , with matrix elements  $C_{ij}$ , which is the inverse of  $S$  [23]. Therefore, it is important to be able to obtain the discontinuities in the elastic stiffness matrix in terms of the elastic compliance matrix. Thus, close to the transition line,  $C(T_c + 0^+) = C(T_c - 0^+) + \Delta C$  and  $S(T_c + 0^+) = S(T_c - 0^+) + \Delta S$ , where  $0^+$  is positive and infinitesimal. By making use of the fact that  $C(T_c + 0^+) S(T_c + 0^+) = \hat{1}$ , where  $\hat{1}$  is the unit matrix, it is shown that, to first order, the discontinuities satisfy,



$\Delta C \approx -C \Delta S C$ . In this manner, it is found that, for instance at  $T_{c+}$ ,  $\Delta C_{11}^+ \approx (2(C_{j1} d_{j1})^2/b_1)$ . From this expression it is clear that  $\Delta C_{11}^+$  must be greater than zero. In general, at  $T_{c+}$ ,  $T_{c-}$ , and  $T_{c0}$ , the expressions that define the jumps for the discontinuities in elastic stiffness and compliances, due to an external stress, have either a positive or a negative value. In this way,  $\Delta S_{11}$ ,  $\Delta S_{22}$ ,  $\Delta S_{33}$ , and  $\Delta S_{66}$  are all negative; while, the stiffness components  $\Delta C_{11}$ ,  $\Delta C_{22}$ ,  $\Delta C_{33}$ , and  $\Delta C_{66}$  are all positive.

## 5. Final remarks

Since for  $\text{Sr}_2\text{RuO}_4$ , the symmetry-breaking field, due to  $\sigma_i$ , is under experimental control, states of zero symmetry-breaking stress and of  $\sigma_i$  single direction can be achieved [1–3]. Hence, it has significant advantages the use of  $\text{Sr}_2\text{RuO}_4$  as a material in detailed studies of superconductivity symmetry-breaking effects, described by a two-component order parameter. Nevertheless, determining from  $\text{Sr}_2\text{RuO}_4$  experimental measurements the magnitude of the parameters in the Ginzburg-Landau model is complicated, because the number of independent parameters occurring for the case of tetragonal symmetry is greater than for the case of hexagonal symmetry (*i.e.*  $UPt_3$ ) [24–26]. Thus for  $\text{Sr}_2\text{RuO}_4$ , three linearly independent parameters,  $b_1$ ,  $b_2$ , and  $b_3$ , are required to specify the fourth order terms in  $\Psi$  occurring in Eq. (1); whereas only two independent parameters,  $b_1$  and  $b_2$ , are required for  $UPt_3$ . For  $\text{Sr}_2\text{RuO}_4$ , two independent ratios

can be formed from the three independent  $b_i$  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  $\sigma_1$  and  $\sigma_6$  [4, 11].

Measurements results for the  $\text{Sr}_2\text{RuO}_4$  elastic constants below  $T_c$  are presented in Ref. 2. There, it is concluded that the quantities  $C_{44}$  and  $C_{11} - C_{12}$  follow the same behavior as those of the BCS superconducting transition, which is evidenced by a change in slope below  $T_{c0}$ . On the other hand, a discontinuity is observed for  $C_{66}$  below  $T_{c0}$ , without a significant change in the sound speed slope as  $T$  goes below 1 Kelvin. It has been previously stated [2, 11] that this kind of  $C_{66}$  changes can be understood as a signature of an unconventional transition to a superconducting phase. Thus, this set of results and others, as those of Clifford *et al.* [3], lead to consider  $\text{Sr}_2\text{RuO}_4$  as an excellent candidate for a detailed experimental investigation of the effects of a symmetry-breaking field in unconventional superconductors.

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- i.* the Voigt notation for  $C_{66}$  means  $C_{xyxy}$  where  $6 = xy$  [15].
  - ii.* The invariance under the gauge symmetry  $U(1)$  means that the quantities  $\psi_i$  must transform according to the rule  $\psi_x \rightarrow e^{i\Phi} \psi_x$  and  $\psi_y \rightarrow e^{i\Phi} \psi_y$ .
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