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Symmetry field breaking effects in Sr_2RuO_4

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In this work, after reviewing the theory of the elastic properties of Sr_2RuO_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 Sr_2RuO_4 superconducting order parameter Ψ 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 Sr_2RuO_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 Ψ 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 Ψ is represented by a three-dimensional vector $\mathbf{d}(\mathbf{k})$. If Ψ 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 Sr_2RuO_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 Ψ breaks time reversal symmetry, which constitutes another key feature of unconventionality.

The Sr_2RuO_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 implies that Ψ has two different components with the time reversal symmetry broken. Similar conclusions from a muon spin relaxation (μSR) experiment were reported by Luke *et al*. [10]. Recently, experiments on the effects of uniaxial stress σ_i, as a symmetry-breaking field were performed by Clifford and collaborators [3], reporting that for Sr_2RuO_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 Sr_2RuO_4 is an unconventional superconductor with a two-component Ψ [4, 11]. Here, let us mention that a different theory of Sr_2RuO_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 σ_i, and directly calculates the jumps at zero stress, where the derivative of T with respect to σ_i doesn't exist.

In this work, we first perform an analysis based on a Ψ that transforms as one of the two dimensional irreducible representations of the Sr_2RuO_4 point group [4, 13]. Subsequently, we construct the Sr_2RuO_4 superconducting phase diagram under an external σ_i. This phase diagram is employed to develop a complete theory of the elastic behavior of Sr_2RuO_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 Sr_2RuO_4 as a material for a detailed study of symmetry-breaking effects in superconductivity described by a two-component Ψ.

2. Ehrenfest relations for a uniaxial stress σ_i

Provided that σ_i does not split the phase transition [4], for applied σ_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 σ_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 σ_i, the elastic strain $e_i = -(\partial G/\partial \sigma_i)_T$ are continuous functions of σ_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

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

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,

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

From the first expression in Eq. (2), the relation for α_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 σ_i produces a phase transition splitting.

3. Ginzburg-Landau model

In this section, a phenomenological model which takes into account the Sr_2RuO_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 Sr_2RuO_4 must be described by a Ψ, 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^Γ, we first will suppose that the Sr_2RuO_4 superconductivity is described by an order parameter ψ^Γ, which transforms according to one of the eight one-dimensional representations of D_{4h}: Γ = 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^Γ linear in σ_i and quadratic in ψ^Γ:

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

The terms proportional to σ_{xx}, σ_{yy} and σ_{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^i_{66} arise from the linear coupling with σ_{xy}. However, due to symmetry, the later linear coupling does not exist for any Γ; 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 ψ^Γ. 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 Sr_2RuO_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 Γ, 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 Γ, the superconductivity in Sr_2RuO_4 must be described by an order parameter ψ^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 Sr_2RuO_4 , and the speed measurements are analyzed in terms of the model $\psi^{\mathrm{E}} = (\psi_x, \psi_y)$, with ψ_x and ψ_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^{Γ}. To maintain gauge symmetry, only real and even products of Ψ can occur in the expansion of G^{Γ}; thus, we find that all real invariants should be formed by second and fourth order products of ψ's^{ii}. 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 ψ'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 σ_i case, the expansion of G gives place to:

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

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 (ψ_x, ψ_y) with the form:

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

where η_x and η_y are both real and larger than zero. After substitution of ψ_x and ψ_y in Eq. (7) , G becomes:

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

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 φ = 0. Moreover, if η_x and η_y have the form $\eta_x = \eta \;\; \sin\chi$ and $\eta_y = \eta \;\; \cos\chi$, G becomes

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

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 η_x = η_y. On the other hand, if $\tilde{b} < 0$, then G becomes minimal if $\sin^2 2\chi$ = 0. In this case, either η_x = 0 or η_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 Sr_2RuO_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 σ_6 and why it gives rise to the discontinuity in S_{66} [11]. Minimization of Eq. (7) with respect to φ and χ, 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 φ and χ. Thus, for the E representation, solutions of the form,

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

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:

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

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 ψ_1, ψ_2 and ψ_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 σ_6 must be taken to be different from zero. If the solution ψ_2 is considered, the term containing σ_6 becomes zero; therefore it is not acceptable. On the other hand, this requirement is satisfied by Ψ_3, with the form $(1, i)\eta$. Hence, the GL analysis renders Ψ_3 as the solution that breaks time reversal symmetry.

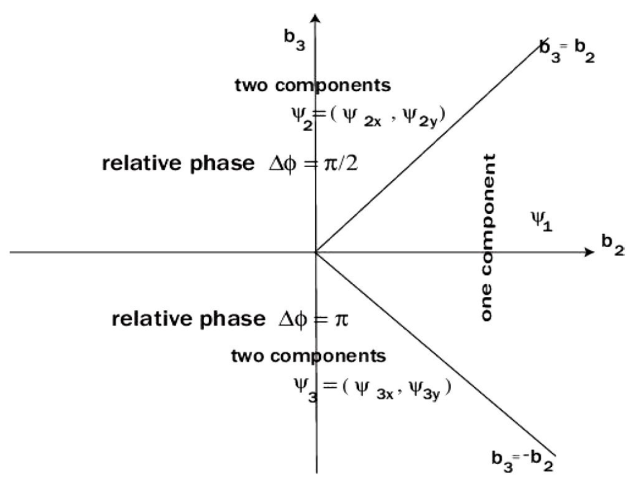

FIGURE 1. Superconducting state phase diagram for the two dimensional representation E of the tetragonal group D_4 as function of the material parameters b_2 and b_3 showing the domains which correspond to the order parameters ψ_1, ψ_2, and ψ_3. Each domain corresponds to a different superconducting 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 Ψ with e_{ij} and in first order Ψ with σ_{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 Sr_2RuO_4 basal plane is given by

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

Here, Λ_i are the temperature-dependent α_i, d_{ij} are the coupling terms between Ψ 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 σ_6 and Ψ. The tensor d_{ij} couples E_i with σ_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, Ψ and σ_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 Ψ 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

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

In what follows, we assume that $d_{11} - d_{12} > 0$, such that $T_{c+} > T_{cy}$. Notice that this does not imply any lost in generality, assuming $d_{11} - d_{12} < 0$, would render an identical model, simply by exchanging the x and y indices. Here, T_{c+} is the higher of the two critical temperatures at which the initial transition occurs. As should be expected, just below T_{c+}, only ψ_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 ψ_y is also different from zero (see Fig. (2)). Thus, in the presence of a non zero compressible σ_1, Ψ has the form $(\psi_x, \psi_y) \approx \psi(1, \pm i\,\epsilon)$, where ϵ 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).

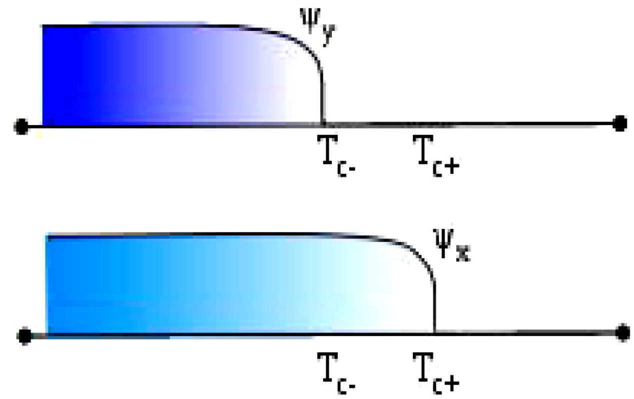

FIGURE 2. Temperature behavior of the two component order parameter (ψ_x, ψ_y) for the case of a nonzero uniaxial stress below T_c. Notice that only the BCS component $\psi_x(T_{c+})$ becomes non zero for temperatures between T_{c+} and T_{c-}. The second unconventional component $\psi_y(T_{c-})$ only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

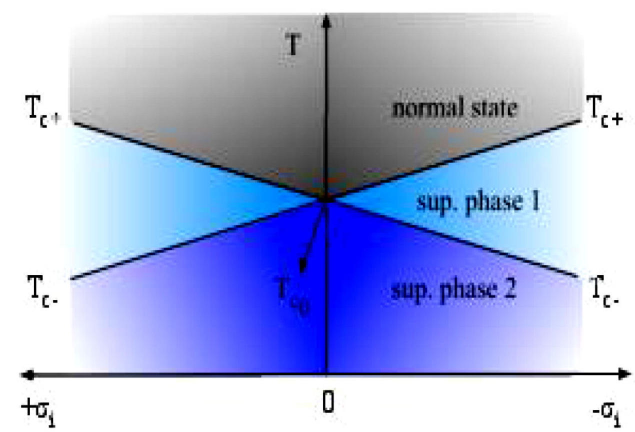

FIGURE 3. Phase diagram showing the upper and lower superconducting transition temperatures, T_{c+} and T_{c-}, respectively, as functions of the compressible stress $-\sigma_i$ along the a axis.

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

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

To obtain Eq. (16), it is assumed that $\sigma_1^2 \ll \sigma_1$ and only linear terms in σ_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_2RuO_4 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_1, the analysis of the behavior of the sound speed at T_c requires a systematic study of these second-order phase transitions. Moreover, thermodynamic quantities, such as $dT_c/d\sigma_i$, C_σ, and α_σ, which are needed in order to calculate the components S^{σ}_{ij} are accompanied by a discontinuity at each of the second order phase transitions.

As depicted in Fig. (3), for a given $\sigma_1 \neq 0$ as T is lowered below T_{c+}, 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, $T = T_{c+}$ (σ_i) is given by $\Delta Q^+ = Q(T_{c+} + 0^+) - Q(T_{c+} - 0^+)$, where 0^+ is a positive infinitesimal number. If T is further dropped below T_{c-}, a second discontinuity arises, and the lower line of transition temperatures appears. The discontinuity along this line, at $T = T_{c-}$ (σ_i), is defined by $\Delta Q^- = Q(T_{c-} + 0^+) - Q(T_{c-} - 0^+)$ [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_{c0}, for the case with σ_i= 0, where the Ehrenfest relations do not hold directly [4]. As an example of these discontinuities, the two jumps in C_σ under an external σ_i are sketched in Fig. (4).

4.1. Jumps due to a uniaxial stress σ_1

The free energy, Eq. (13), for the cases where both σ_1 and σ_6 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 $\alpha_x = \alpha'(T - T_{c0}) + \sigma_1 d_{11}$ and $\alpha_y = \alpha'(T - T_{c0}) + \sigma_1 d_{12}$. If only σ_1 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 $\Delta G = G - G_0(T)$. The nature of the superconducting state that follows from Eq. (19), depends on the values of the coefficients b_1, b_2, and b_3. 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_{c+} and in the presence of σ_1, the second order terms in Eq. (19) dominate and Ψ has a single component ψ_x; whereas at T_{c-} a second component ψ_y 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_2 given by Eq. (12). In this case, G can be written in terms of η_x and η_y 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_1, b_2, and b_3. Assuming that $b_3 > 0$, and η_x and η_y 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 $\epsilon = 0$ to $\epsilon \approx 1$ as T is reduced below T_{c-}, 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_{c+}, we use $\alpha_x = \alpha'(T - T_{c+})$ and $\alpha_y = \alpha'(T - T_{cy})$, and assume that $T_{c+} > T_{cy}$. For the interval $T_{c+} > T > T_{c-}$, the equilibrium value for Ψ satisfies $\alpha_x > 0$ and $\alpha_y = 0$, *i.e.* $\eta_x > 0$ and $\eta_y = 0$, with $\eta_x^2 = -2\alpha_x/b_1$, obtaining that T_{c+} and its derivative with respect to σ_1 are respectively,

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

The specific heat discontinuity at T_{c+}, relative to its normal state value, is calculated by using:

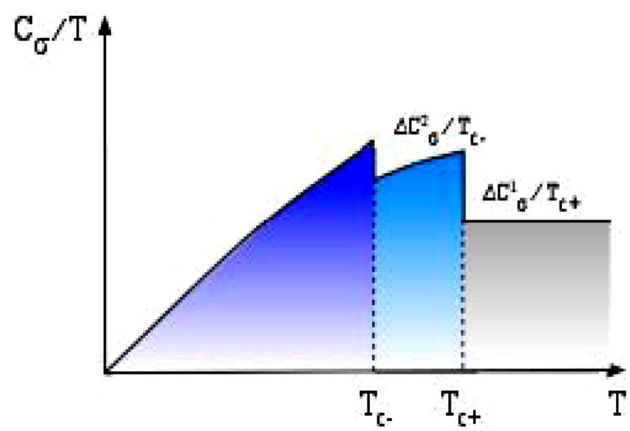

FIGURE 4. Schematic dependence of the specific heat on the temperature, for the case of an uniaxial stress splitting the Sr_2RuO_4 transition Temperature. Notice the two jumps in the heat capacity near the transition temperatures T_{c+} and T_{c-}.

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

and renders the result

$$\Delta C^{+}_{\sigma_1} = -\frac{2\ T_{c+}\alpha'^2}{b_1}. \tag{25}$$

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

$$\Delta\alpha^{+}_{1} = -\frac{2\alpha'\ d_{11}}{b_1}. \tag{26}$$

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

$$\Delta S^{+}_{i'j'} = -\frac{2\ d_{i'1}\ d_{j'1}}{b_1}. \tag{27}$$

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 $\Delta S^{\sigma_1}_{11} = -\frac{2\ d^2_{11}}{b_1}$.

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,

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

In this expression, the second order term in η_y is renormalized by the square of η_x. The second transition temperature is determined from the zero of the total prefactor of η_y^2, obtaining that T_{c-} and its derivative with respect to σ_1 are:

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

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

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

This analysis shows that the second superconducting phase is different in symmetry, and that time reversal symmetry is broken. The change in C_{σ_1} at T_{c-}, with respect to its value in the normal phase, $\Delta C^{-,N}_{\sigma_1}$, is found to be, $\Delta C^{-,N}_{\sigma_1} = -2\ T_{c-}\alpha'\ 1/b$. The specific heat variation at T_{c-} is,

$$\Delta C^{-}_{\sigma_1} = \Delta C^{-,N}_{\sigma_1} - \Delta C^{+}_{\sigma_1}, \tag{31}$$

which results in

$$\Delta C^{-}_{\sigma_1} = -2\ T_{c-}\ \alpha'^2\frac{\tilde{b}}{b\ b_1}. \tag{32}$$

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 α_i at T_{c-} is obtained to be

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

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

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

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:

$$\Delta C^{0}_{\sigma_1} = -\frac{2T_{c0}\alpha'^2}{b}, \quad \Delta\alpha^{0}_{i'} = -\frac{\alpha' d_{i'+}}{b},$$
$$\Delta S^{0}_{i'j'} = -\frac{1}{2}\left(\frac{d_{i'+}\ d_{j'+}}{b} + \frac{d_{i'-}\ d_{j'-}}{\tilde{b}}\right). \tag{35}$$

Before continuing, it is important to emphasize that at at zero stress, the derivative of T_c with respect to σ_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 σ_6

When a shear stress σ_6 is applied to the basal plane of Sr_2RuO_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 σ_6 *i.e.* $dT_c/d\sigma_6$ is different for each of the two transition lines. At each of these transitions, C_{σ_6}, α_{σ_6}, and $S^{\sigma_6}_{ij}$ 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 σ_1; therefore, the diagram in Fig. (3) also qualitatively holds here. In the case of an applied σ_6, ΔG is given by

$$\Delta G = \alpha(|\psi_x|^2 + |\psi_y|^2) + \sigma_6 d_{66}(\psi_x\psi_y^* + \psi_x^*\psi_y) + \frac{b_1}{4}(|\psi_x|^2 + |\psi_y|^2)^2 + b_2|\psi_x|^2|\psi_y|^2 + \frac{b_3}{2}(\psi_x^2\psi_y^{*2} + \psi_y^2\psi_x^{*2}). \tag{36}$$

Here $\alpha = \alpha'(T - T_{c0})$, and the minimization of ΔG is performed as in the σ_1 case, *i.e.* by substituting the general expression for Ψ given in Eq. (8). After doing so, ΔG becomes

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

In the presence of σ_6, the second order term determines the phase below T_{c+}, which is characterized by ψ_x and by $\psi_y = 0$. As the temperature is lowered below T_{c-}, depending of the value of b$_3$ a second component ψ_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 $\sigma_6 \to 0$, 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 η_x and η_y, and of the phase φ. If $b_3 < 0$, and η_x and η_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

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

ΔG 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 σ_6 is non zero, the state with the lowest free energy corresponds to $b_3 < 0$, phase φ equal to $\pi/2$, and Ψ of the form:

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

In phase 1 of Fig. (3), $\varphi = 0$, and as T is lowered below T_{c-}, phase 2, φ grows from 0 to approximately $\pi/2$. Again, following an analysis similar to that carried out for σ_1, the two transition temperatures T_{c+} and T_{c-} are obtained to be:

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

The derivative of T_{c+} with respect to σ_6, and the discontinuity in $C^+_{\sigma_6}$ at T_{c+} are respectively found to be:

$$\frac{d\,T_{c+}}{d\,\sigma_6} = -\frac{d_{66}}{\alpha'}, \qquad \Delta C^+_{\sigma_6} = -\frac{2\,T_{c+}\,\alpha'^2}{b'}. \quad (42)$$

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

$$\Delta\alpha^+_{\sigma_6} = -\frac{2\,\alpha'\,d_{66}}{b'}, \qquad \Delta S^+_{66} = -\frac{2\,d_{66}^2}{b'}. \quad (43)$$

For T_{c-}, the derivative of this transition temperature with respect to σ_6, and the discontinuities in the specific heat, thermal expansion and elastic stiffness respectively are:

$$\frac{d\,T_{c-}}{d\,\sigma_6} = \frac{b\,d_{66}}{2\,b_3\,\alpha'}, \qquad \Delta C^-_{\sigma_6} = -\frac{4\,T_{c-}\,\alpha'^2\,b_3}{b\,b'}, \quad (44)$$

$$\Delta\alpha^-_{\sigma_6} = \frac{2\,\alpha'\,d_{66}}{b'}, \qquad \Delta S^-_{66} = -\frac{d_{66}^2\,b}{b'b_3}. \quad (45)$$

Since for the case of σ_6, the derivative of T_c with respect to σ_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 σ_6, are calculated by adding the expressions obtained for the discontinuities at T_{c+} and T_{c-},

$$\Delta C^0_{\sigma_6} = -\frac{2\,T_{c0}\,\alpha'^2}{b}, \qquad \Delta S^0_{66} = -\frac{d_{66}^2}{b_3}, \qquad \Delta\alpha^0_{\sigma_6} = 0. \quad (46)$$

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

Since the phase diagram was determined as a function of σ_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 6×6 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 ΔC^{+}_{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, ΔS_{11}, ΔS_{22}, ΔS_{33}, and ΔS_{66} are all negative; while, the stiffness components ΔC_{11}, ΔC_{22}, ΔC_{33}, and ΔC_{66} are all positive.

5. Final remarks

Since for Sr_2RuO_4, the symmetry-breaking field, due to σ_i, is under experimental control, states of zero symmetry-breaking stress and of σ_i single direction can be achieved [1–3]. Hence, it has significant advantages the use of Sr_2RuO_4 as a material in detailed studies of superconductivity symmetry-breaking effects, described by a two-component order parameter. Nevertheless, determining from Sr_2RuO_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 Sr_2RuO_4, three linearly independent parameters, b_1, b_2, and b_3, are required to specify the fourth order terms in Ψ occurring in Eq. (1); whereas only two independent parameters, b_1 and b_2, are required for UPt_3. For Sr_2RuO_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 σ_1 and σ_6 [4, 11].

Measurements results for the Sr_2RuO_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 Sr_2RuO_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 ψ_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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