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Ginzburg-Landau theory and effects of pressure on a two-band superconductor: application to MgB_2

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Abstract. We present a model of pressure effects of a two-band superconductor based on a Ginzburg-Landau free energy with two order parameters. The parameters of the theory are pressure as well as temperature dependent. New pressure effects emerge as a result of the competition between the two bands. The theory then is applied to MgB_2. We identify two possible scenaria regarding the fate of the two σ subbands under pressure, depending on whether or not both subbands are above the Fermi energy at ambient pressure. The splitting of the two subbands is probably caused by the E_{2g} distortion. If only one subband is above the Fermi energy at ambient pressure (scenario I), application of pressure diminishes the splitting and it is possible that the lower subband participates in the superconductivity. The corresponding crossover pressure and Grüneisen parameter are estimated. In the second scenario both bands start above the Fermi energy and they move below it, either by pressure or *via* the substitution of Mg by Al. In both scenaria, the possibility of electronical topological transition is emphasized. Experimental signatures of both scenaria are presented and existing experiments are discussed in the light of the different physical pictures.

PACS. 74.20.De Phenomenological theories (two-fluid, Ginzburg-Landau, etc.) – 74.25.Dw Superconductivity phase diagrams – 74.62.Fj Pressure effects – 74.70.Ad Metals; alloys and binary compounds (including A15, MgB2, etc.)

1 Introduction

The discovery of superconductivity in the material MgB_2 [1] initiated intensive recent theoretical and experimental interest. The possibility of a high critical temperature in a class of materials which are chemically much simpler than the high-T_c cuprates and the occurence of large critical current densities pose some interesting new questions in the research of superconductivity. MgB_2 is a type II superconductor in the clean limit [2]. The crystal belongs to the space group P6/mmm or AlB_2-structure where borons are packed in honeycomb layers alternating with hexagonal layers of magnesium ions. The ions Mg^{2+} are positioned above the centers of hexagons formed by boron sites and donate their electrons to the boron planes. The electronic structure is organized by the narrow energy bands with near two-fold degenerate (σ-electrons) and the wide-band (π-electrons) [3–5]. Without any of the lattice strain, the σ dispersion relations are slightly splitted due to the two boron atoms per unit cell (the electronic analog of Davydov splitting for phonons). The σ portions of the Fermi surface (FS) consist of coaxial cylinders along the Γ-A symmetry direction of the Brillouin zone (BZ), whereas the π-bands are strongly dispersive.

The excited phonon modes in MgB_2 present a sharp cut-off at about 100 meV. The optical modes (B_{1g}, E_{2g}, A_{2u} and E_{1u}) are practically non-dispersive, along the Γ-A direction. Various Raman data [6–8] show a small spread of the E_{2g} frequency around 74.5 meV in different ceramic samples of MgB_2. The strong deformation potential Δ of the in-plane E_{2g} mode [9,10] causes a significant energy splitting of the σ-band around 1.5 eV, lifting its two-fold degeneracy at the Γ-point of the BZ. In general, measurements of the effect of pressure on the electronic structure are based on the Raman technique, therefore only the influence on the E_{2g} mode can be traced. High pressure experiments up to 15 GPa [6] have revealed a large increase of the out of phase E_{2g} phonon mode. This leads to a suppression of the displacement of the boron atoms as well as of the deformation potential $\Delta = B_2u^2 + B_4u^4$ where u is the displacement [10] which will be used below. Moreover, experiments under pressure up to 40 GPa do not show any structural phase

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transition [6,11–13]. Due to the anisotropy the hydrostatic pressure decreases the inter-plane distance more than the in-plane distance between adjacent borons.

The interpretation of experimental data from different spectroscopic methods [14–16], suggests the presence of two different superconducting gaps. The specific heat behavior [17], the low isotope effect [18], pressure effects [19] and penetration depth data [20] provide evidence for a complicated superconducting order parameter in MgB_2. Recent NMR measurements [21] of ^{11}B as well as Hall measurements [22] are consistent with a σ-band driven superconductivity in MgB_2 where the π-band participates due to interband scattering. The use of two order parameters is justified by recent experimental results on scanning tunneling microscopy [23].

Some microscopic theories [9,24] are based on σ-band or σ-π band scenaria for superconductivity whereas others [25] are concentrated on a π-band superconductivity. A consensus is formed though, that the driving band for the superconductivity is the σ one. The calculated spectral functions [26] and the analysis of the reflectance measurements [27] show the possibility of different superconducting mechanisms beyond the conventional electron-phonon Bardeen-Cooper-Schrieffer (BCS) pairing. The complicated nature of superconductivity in MgB_2 does not allow at present to accept the conventional superconducting mechanism. Therefore, it is appropriate to construct a phenomenological theory.

Since MgB_2 presents an example of a two-band superconductor [28] may serve as a paradigm to investigate new effects. The purpose of this article is to present a phenomenological Ginzburg-Landau (GL) theory for a two-band superconductor appropriate to MgB_2 based on two order parameters and, in that framework, to provide a physical picture of pressure effects on MgB_2. The study of pressure effects is one way to investigate (i) the different topology of the two bands and (ii) the question of the participation of the two σ subbands in the superconductivity. A brief account of a part of this work was presented elsewhere [29]. In the present article missing terms in the GL functional are restored, non-adiabatic effects are explained using a different approach and the effects of pressure are discussed in more detail and compared with very recent experiments. The work is organized as follows. In Section 2 the GL description is presented, in Section 3 the pressure effects, in connection with the σ-band, are discussed and a general discussion with some concluding remarks are given in Section 4.

2 Ginzburg-Landau description

We introduce the GL free energy functional with two order parameters, appropriate for MgB_2, assuming that both the order parameters belong to the Γ_1 representation of the point group of MgB_2 crystal. The two order parameters are labeled by the corresponding band (σ or π) of MgB_2, without loss of generality:

$$F = \int \mathrm{d}^3 r \Big\{ \frac{1}{2m_\sigma} |\boldsymbol{\Pi}\psi_\sigma|^2 + \alpha_\sigma |\psi_\sigma|^2 + \beta_\sigma |\psi_\sigma|^4 + \frac{1}{2m_\pi} |\boldsymbol{\Pi}\psi_\pi|^2 + \alpha_\pi |\psi_\pi|^2 + \beta_\pi |\psi_\pi|^4 + r(\psi_\sigma^* \psi_\pi + \psi_\sigma \psi_\pi^*) + \gamma_1 (\Pi_x \psi_\sigma \Pi_x^* \psi_\pi^* + \Pi_y \psi_\sigma \Pi_y^* \psi_\pi^* + \text{c.c.}) + \beta |\psi_\sigma|^2 |\psi_\pi|^2 + \frac{1}{8\pi} (\boldsymbol{\nabla} \times \boldsymbol{A})^2 \Big\}, \quad (1)$$

where $\boldsymbol{\Pi} = -\mathrm{i}\hbar\boldsymbol{\nabla} - 2e/c\boldsymbol{A}$ is the momentum operator, $\boldsymbol{A}$ is the vector potential and $\alpha_{\sigma,\pi} = \alpha^0_{\sigma,\pi}(T - T^0_{c\sigma,\pi})$. It is possible to take into account the large anisotropy of the two order parameters (the σ is almost two-dimensional) by rescaling of the axis according to the effective masses (directional dependence effective mass). In the present work, since we are focusing exclusively on pressure effects, the derivative terms are not important.

In equation (1) we have used the fact that the mixing r-term favors the coupling of linear combination of the two order parameters with phase difference 0 if $r < 0$ or π if $r > 0$ between them [30], therefore a term $\psi_\sigma^{2*}\psi_\pi^2 + \text{c.c.}$ is incorporated into the β-term of the free energy [31]. If $r = 0$ then equation (1) is the free energy for two bands without Josephson coupling between them. The quartic β-term is the only one which mixes the two gaps and is unimportant in this case. The onset of the superconducting state in one band does not imply the onset in the other. This corresponds to the case of $V_{sd} = 0$ in the two-band BCS treatment [32]. If $r \neq 0$, which is the case of MgB_2, then the pair transfering term is present in the GL functional and it means that the onset of superconductivity in one band implies automatically the appearance of superconductivity in the other. There is a single observed T_c which is a function of the bare ones $T^0_{c\sigma,\pi}$. In MgB_2 the compression due to pressure is anisotropic [6,33,34]. According to reference [6] the compressibility along the c-axis is almost twice larger than that of the plane compressibility. Therefore application of uniaxial pressure on single crystals will affect differently the two gaps. Following Ozaki's formulation [35,36], we may add to the GL functional, equation (1), the term which couples the order parameters, in second order, with the strain tensor ϵ to first order, having already specified the symmetry of the order parameters:

$$F_{strain} = -C_1(\Gamma_1)[\delta(\epsilon_{xx} + \epsilon_{yy}) + \epsilon_{zz}]|\psi_\sigma|^2 - C_2(\Gamma_1)(\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz})|\psi_\pi|^2 - C_3(\Gamma_1)[\delta'(\epsilon_{xx} + \epsilon_{yy}) + \epsilon_{zz}](\psi_\sigma^* \psi_\pi + \text{c.c.}), \quad (2)$$

where $C_{1,2,3}(\Gamma_1)$ are coupling constants and δ, δ' are given in terms of the elastic constants of the material. This will lead to a change of the bare critical temperatures $T^0_{c\sigma,\pi}$ and consequently of the actual critical temperature T_c. The physical reasoning behind this is that the material will change in such a way as to gain condensation energy by enhancing the density of states in the direction where

the superconducting gap is larger. The term proportional to $\psi_\sigma^*\psi_\pi$ + c.c. in the F_{strain} will renormalize the mixing coupling r to $\tilde{r}$. We model the corresponding differences by taking a linear dependence of the two bare T_c's with pressure $T_{c\sigma,\pi} = T^0_{c\sigma,\pi} - \eta_{\sigma,\pi}p$, where $\eta_{\sigma,\pi} = |\partial T^0_{c\sigma,\pi}(p = 0)/\partial p|$ and we will discuss the validity of the assumption for the MgB_2 later.

Analyzing the equations which result from the minimization of the GL free energy we get:

$$\alpha_\pi \alpha_\sigma = \tilde{r}^2. \tag{3}$$

This gives the pressure dependence of the superconducting critical temperature:

$$T_c(p) = \frac{1}{2}\left[T^0_{c\sigma} + T^0_{c\pi} - (\eta_\pi + \eta_\sigma)p\right] + \frac{1}{2}\left\{[T^0_{c\sigma} - T^0_{c\pi} + (\eta_\pi - \eta_\sigma)p]^2 + a^2\right\}^{1/2}, \tag{4}$$

where $a^2 = 4\tilde{r}^2/(\alpha^0_\pi \alpha^0_\sigma)$. Deviations from a straight line at moderate values of pressure can be attributed to the two bands. From the above formula the inequality $\mathrm{d}T_c/\mathrm{d}p < 0$ is always true as long as the renormalized mixing parameter $\tilde{r}$ is real, as a consequence of the initial assumption on the pressure dependence of the bare critical temperatures. In the case of MgB_2 we can safely consider that $\eta_\pi > \eta_\sigma$.

3 Nonadiabatic effects

We now wish to address an issue specific to MgB_2 which can be also realized in other multiband superconductors. The physical situation we would like to question is the splitting of the two σ subbands at ambient pressure and their participation in superconductivity with the increase of pressure. Due to contradictory experimental as well as theoretical results we proceed by distinguishing two different scenaria. The first one addresses the splitting of the two sigma subbands at ambient pressure as shown by first princible calculations [9,10,37] when one of the subbands is below the Fermi energy E_F. The second scenario addresses the opposite situation where both the σ subbands are above E_F at ambient pressure.

3.1 Scenario I: one σ sub-band above E_F at ambient pressure.

In particular in [37] the splitting of the two subbands was studied in detail and compared with similar situation in AlB_2. The conclusion is that due to the E_{2g} phonon mode, the two σ bands split nearby Γ point and the lower band completely sinks below the Fermi energy. This is the case for MgB_2 and also for the heavily hole-doped graphite. Moreover, experimental results [33,38] showed a kink in the superconducting critical temperature as a function of pressure at approximately 6–8 GPa and also a kink in the volume dependence of T_c for $Mg^{10}B_2$ at around 20 GPa and $Mg^{11}B_2$ at around 15 GPa [7].

The physics we discuss here, is the change in the electronic properties of the material under pressure and its influence on the superconducting state. The band which is below the Fermi level at ambient pressure [9] is possible to overcome the energy difference and to get above the Fermi level at a certain value of pressure (termed as crossover pressure), restoring the degeneracy of the two σ-bands at point Γ. This is a non-adiabatic effect as discussed in [37]. To understand and illustrate this effect on the superconducting state we need to consider the pressure dependence of the coefficient α_σ as usual [39] with the modification due to the particular physical situation by writing:

$$\alpha_\sigma = \alpha^0_\sigma(T - T^0_{c\sigma} + \eta_\sigma p) + \alpha^1_\sigma(p - p_c)\Theta(p - p_c), \tag{5}$$

where $\Theta(x)$ is the Heaviside step function, α^0_σ and α^1_σ are positive constants. This choice reflects the fact that at a certain crossover pressure p_c the second band starts to participate in superconductivity as well and that initially it is an increasing function of pressure. We obtained the above formula by requiring the continuity of the coefficient α_σ at p_c.

After the energy difference between the two σ-bands almost disappears, then they both follow the same reduction of the bare T_c's with pressure. The approximate value of the crossover pressure can be estimated as follows. The energy difference between the two subbands is approximated by $\delta E \simeq (1-n)\sqrt{\Delta}$, where the fraction of superconducting electrons is $1-n \sim 0.03$ [3], *i.e.* the carrier density per boron atom in MgB_2. The approximate value of p_c is half the value of the pressure which suppresses the deformation potential Δ. The deformation potential in turn, can be estimated by the expression $p_c\Omega \sim (1-n)\sqrt{\Delta}$ where $\Omega \sim 30$ Å^3 is a unit cell volume of MgB_2 and $\Delta = 0.04$ eV^2 is the deformation potential for a boron displacement $u \sim 0.03$ Å [10]. Using these parameters, we get a crossover pressure of $p_c \sim 30$ GPa. This estimate shows, that a realistic applied pressure influences drastically the electronic structure and the FS topology, restoring the degeneracy of the two subbands at the Γ-point which are initially splitted. Superconductivity in the second subband may occur at lower values of the estimated p_c due to the fact that both subbands are affected. The elimination of the energy difference around the Fermi energy will also occur at lower values due to the corrugation of the σ portions of the FS, the already existing strains in the material and the anisotropic compressibility. These considerations make the above estimate an upper limit of the crossover pressure p_c. The degree to which shear stresses of the sample and the surrounding fluid under pressure affect the data of pressure measurements is still under investigation [19].

The microscopic nature of this crossover is caused by the change of the FS. If the topology of the FS changes under pressure (this subsection) or due to substitutions in the composition (next subsection), a van Hove singularity at critical energy E_c in the electronic density of states $\rho(E)$ is manifested in an electronic topological transition of the 5/2 kind [40]. Since $\mathrm{d}T_c/\mathrm{d}p$ is

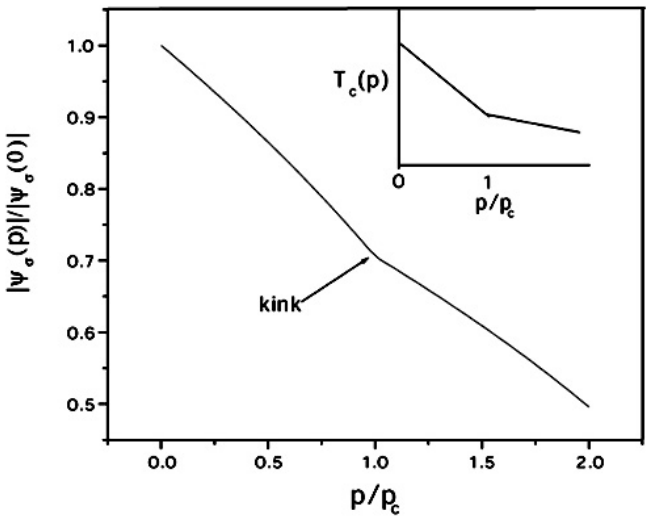

Fig. 1. The form of the order parameter as a function of pressure, where the kink at p_c is shown. Inset: the behavior of the critical temperature $T_c(p)$. The chosen parameters are (in reduced units): $T = 0.8T^0_{c\sigma}$, $T^0_{c\sigma} = 0.7\ T^0_{c\sigma}$, $\eta_\sigma = 0.2\ T^0_{c\sigma}/p_c$, $\eta_\pi = 0.5\ T^0_{c\sigma}/p_c$, $\alpha^0_\sigma = 2$, $\alpha^0_\pi = 1$, $\alpha^1_\sigma = 0.1$.

essentially proportional to the derivative $\mathrm{d}\rho(E_F)/\mathrm{d}E_F$, it develops singularities $\propto 1/\sqrt{E_F - E_c}$, provided that the strength of carrier attraction varies slightly with pressure [41].

In Figure 1 using a model calculation, we illustrate the expected behavior of T_c and the form of the order parameter as a function of pressure at fixed temperature and, schematically, the kink of T_c at p_c due to the change of the slope from equation (5). The predicted kink close to p_c can be detected directly in a penetration depth experiment under pressure. The chosen parameters are such that they respect the relation $\eta_\pi > \eta_\sigma$, the fact that the superconducting density coming from the σ band is higher than the one coming from the π-band ($\alpha_\sigma > \alpha_\pi$) and that the driving band for superconductivity is σ ($T^0_\sigma > T^0_\pi$).

The contribution on the Grüneisen parameter:

$$G = B\frac{\mathrm{d}\ln\omega}{\mathrm{d}p} = \frac{B}{\omega}\frac{\mathrm{d}\omega}{\mathrm{d}p}, \tag{6}$$

where B is the bulk modulus and ω is the phonon frequency, can be approximately obtained if we observe that $\delta E \simeq \hbar\omega_{E2g}$ then $G \simeq B/2p_c$. The bulk modulus is measured to be B $\simeq$ 114 GPa [42] and our estimate for the Grüneisen parameter is G $\simeq$ 3.8. Experimentally G $= 2.9 \pm 0.3$ as reported in [6] for the measured Raman active E_{2g} phonon mode. However, as indicated in reference [6], for anisotropic crystals it would be more appropriate to scale the frequency shift in the Grüneisen parameter with the variation of the lattice constant a such as $G = \mathrm{d}ln\omega/3\mathrm{d}lna$ [43]. Then, the Grüneisen parameter for the MgB_2 takes the value 3.9 ± 0.4. There is an excellent agreement with the above estimate and it justifies our approach.

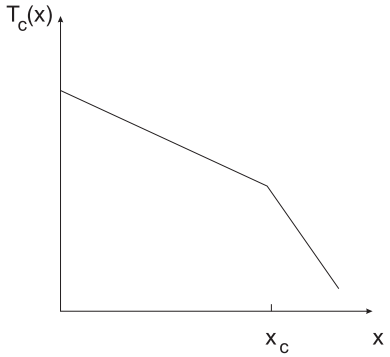

Fig. 2. Schematically the form of the superconducting critical temperature $T_c(x)$ in the second scenario. x is the concentration of Al in a $Mg_{1-x}Al_xB_2$-like system.

3.2 Scenario II: both σ sub-bands above E_F at ambient pressure.

Some recent experimental and theoretical studies [44] lend support to a second scenario. More specifically, the substitution of Mg by the higher valent Al was used to provide the necessary change of the Fermi energy *via* electron concentration and the lattice parameters in the compound $Mg_{1-x}Al_xB_2$. This is in essence equivalent to a change of Fermi surface topology as in the pressure experiments. As it was shown in [44] by increasing the Al content (equivalently by increasing the pressure), a topological crossover occurs at some point and the negative slope of T_c as a function of Al content x decreases further giving a different picture from that of Figure 1.

To model this in the language of the GL model, one basically has to substitute the pressure p and the crossover pressure p_c with the Al concentration x and and the crossover concentration x_c respectively, requiring again continuity at x_c:

$$\alpha_\sigma = \alpha^0_{\sigma,x}(T - T^0_{c\sigma} + \eta_{\sigma,x}x) + \alpha^1_{\sigma,x}(x - x_c)\Theta(x - x_c) \tag{7}$$

where the extra substrcipt x denotes that the numerical coefficients are different from equation (5). The critical temperature as a function of the Al content is illustrated in Figure 2. The physics is similar to the topological transition described in scenario I where now instead of pressure, the changing parameter is the composition. It is a more controlled way to change the topology of the FS. In fact, an abrupt topological change in the σ-band Fermi surface was found at $x = 0.3$ in reference [44]. When the σ bands are filled in $Mg_{1-x}Al_xB_2$ at $x \approx 0.6$, superconductivity dissapears. It is worth noticing that in this compound, the impurity scattering broadens the van Hove singularities at the saddle points E_c of the FS and smears the change of slope of T_c at E_c. Therefore the kink of T_c in Figure 2 will be less pronounced (see also the experiment in Ref. [44]).

We emphasize here the need of de Haas-van Alphen (dHvA) data for $Mg_{1-x}Al_xB_2$, especially close to $x = 0.3$. The pressure derivative of the superconducting critical

temperature as a function of the concentration $\mathrm{d}T_c(x)/\mathrm{d}p$ can be also conclusive since it can clarify the correlation between pressure, Al concentration and the kink at x_c.

4 Discussion

Interestingly enough, there is an obvious discrepancy from the two pictures of the previous section, which calls for more experimental data. Observation of quantum oscillations in dHvA experiments provides the information about the bands, the effective mass of the carriers and the shape of the Fermi surface in MgB_2. In reference [45], *only* the smaller cylindrical Fermi surface along the c-axis was observed (with a hole band mass -0.251 m_e, where m_e is the electron mass). The same authors reported the observation of *only* the larger of the two σ tube-like Fermi surfaces (with a band mass -0.553 m_e) in reference [46]. Although the estimated parameters of the MgB_2 are in agreement with the LDA band calculations [47] future dHvA experiments should clarify the detailed observation of the σ portions of the FS. Also note that angle resolved photoemission spectroscopy experiments [48] detected *one* band along the $\Gamma - \mathrm{K}$ symmetry line of the BZ.

There are a few points in order to discuss further with respect to applicability of the theory developed here. Early pressure experiments [13] demonstrated the overall decrease of T_c with a pressure increase which was attributed to the loss of holes. In two of the samples of [13] there is a linear dependence of T_c on pressure and in two others a weak quadratic dependence. We stress that the samples were polycrystalline and the experiment is effectively under hydrostatic pressure. Also the degree of nonstoichiometry was not known. The almost linear dependence for a wide range of pressures, makes the GL functional as presented, valid for MgB_2.

Experiments on single crystals will be able to clarify the effects which are described. We do not attempt at the moment any actual detailed fitting of experimental data, except from the illustrative fitting in Section 3, because there is no experimental consensus on the different values of key parameters of the theory (*e.g.*, there is a wide range of published data on the value of $\mathrm{d}T_c/\mathrm{d}p$ [19]). More detailed Raman data with optical reflectivity, specific heat and thermal conductivity measurements under pressure in the superconducting phase are needed as well as a penetration depth experiment which can reveal the pressure dependence of the superfluid density. Also the experiments suggested in scenaria I and II will be crucial in the question regarding the topological transition, where E_F can be varied experimentally either by external pressure (Sect. 3.1) or by alloying (Sect. 3.2). Recent Raman spectra at high pressure [38] reveal a reduction of the slope of the pressure-induced frequency shift by about a factor of two, at about 18 GPa which supports the suggestion that MgB_2 may undergo a pressure-induced topological electronic transition. In connection with the appearance of superconductivity under pressure, we stress the high pressure superconducting phase of $CaSi_2$ which superconducts above an applied pressure of 12 GPa with $T_c \sim 14$ K [49].

One proposed experiment which can potentially show the different role of the two bands (σ and π) is the detection of the Leggett mode or the internal Josephson current [50]. Moreover an upward curvature in H_{c2} is also known to be a signature of multicomponent superconductivity.

In summary, we present an analysis of pressure effects, within the GL theory, of a two-band superconductor and apply it to MgB_2. We make predictions for non-adiabatic effects and discuss different experiments from which crucial information can be extracted on the physics of the two participating bands σ and π as well as the more delicate questions on the two σ subbands.

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