

Pressure effects and large polarons in layered MgB₂ superconductor

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We consider the dependence of the MgB₂ superconducting critical temperature on the pressure. Our model exploits the influence of the large polarons on the band structure of the layered MgB₂ superconductor. Namely, the hole Pekar-Fröhlich polarons form quasi-two-dimensional potential wells in the boron plane that shift the positions of the σ and π bands. This energy shift depends on the pressure, and the Cooper pairing of the correlated σ electrons occurs inside polaron wells. The results obtained are as follows: $dT_c/dp \approx -\alpha(5.2 \pm 0.9)$ K/GPa or $dT_c/dp \approx -\alpha(6.9 \pm 1.1)$ K/GPa for a different choice of the Grüneisen parameter. When compared with known experimental data, these give results a reasonable interval for the value of the Fröhlich electron-phonon coupling constant: $\alpha \approx 0.15-0.45$.

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I. INTRODUCTION

Soon after the discovery of superconductivity in MgB₂ the pressure influence on the critical temperature T_c was experimentally studied. The pressure derivatives dT_c/dp were extracted from obtained data. At low pressure ($p \leq 2$ GPa) the experimental results are as follows: $dT_c/dp = -1.2$ K/GPa in Refs. 1 and 2 and -2 K/GPa in Refs. 1, 3, and 4. Experiments at high pressure give the same sign of the pressure derivatives but demonstrate lower values. Namely, $dT_c/dp = (-0.8, -1.07, -1.11)$ K/GPa in Refs. 5, 1, and 6, respectively. Many conventional low- T_c superconductors also reveal the same sign, $dT_c/dp < 0$, but their absolute values are much lower.

Boron ions in crystalline MgB₂ are packed in honeycomb layers alternating with hexagonal layers of magnesium ions. The in-plane distance, $b = 1.78$ Å, between the boron cations, is less than that between magnesium anions, $a = 3.08$ Å. The space separating the boron planes is of the size $c = 3.52$ Å. The magnesium ions are positioned above the centers of hexagons formed by boron sites and donate their electrons to the boron planes: $\text{Mg}^{2+}[\text{B}^-(p^2)]_2$. These p electrons form σ and π bands and the charged Mg layers lift the threefold degeneracy between σ and π electrons and shift the electronic energy bands so that the π band becomes lower than the σ band. The latter crosses the Fermi level, providing the light- and heavy-hole formation. The electronic structure is formed by the narrow energy band of the twofold-degenerate σ electrons and the wideband of the π electrons.

As to the phonon modes in MgB₂, they have a sharp cutoff at about 100 meV. Below this energy the existence of different phonon modes is presumed.⁷ Note that the phonon energies mentioned in Ref. 7 could be overestimated because the approximation used to derive them is valid only for cubic lattices.^{8,9} The anharmonic in-plane breathing mode E_{2g} (74.5 meV) strongly interacts with the narrow-band σ electrons.¹⁰ Due to this fact its energy is lower than that of the out-of-plane tilting boron mode B_{1g} (87.1 meV). In addition, there are low-energy acoustic modes, and indications

are also found¹¹⁻¹³ of the existence of phonon modes with extremely low energies.

Most of the theoretical speculations on the T_c -pressure relationship in MgB₂ are based on semiphenomenological approaches and do not involve microscopic arguments. Also a Ginzburg-Landau approach to the study of the pressure effects has been proposed recently in Ref. 14. In the scope of conventional BCS theory, dT_c/dp is governed by the competition between a decrease of the density of electronic states at the Fermi level and an increase of the Debye frequency in the BCS formula for T_c . The weak electron-phonon coupling constant¹⁵ in MgB₂ and the absence of the peak in the electronic density of states at the vicinity of the Fermi energy¹⁶ cannot provide the BCS scenario.

On the other hand, spectroscopic measurements of the superconducting gap,¹⁷⁻¹⁹ specific heat behavior,^{20,21} the low isotope effect^{22,23} (under the substitution of both B and Mg isotopes), and pressure effects²⁴ point to the likelihood of the complex mechanism of the superconductivity in MgB₂. The calculated spectral function²⁵ and the analysis¹⁵ of the reflectance measurements²⁶ show the possibilities of different superconducting mechanisms beyond the conventional electron-phonon BCS pairing.

A positive pressure derivative of T_c was found^{27,28} in the microscopic model of the hole dressed superconductivity, which opposes the experimental data.

Among optical modes in MgB₂ the E_{2g} mode is the only Raman-active mode. A high-pressure (up to 15 GPa) Raman experiment²⁹ has revealed a large pressure shift of the E_{2g} -phonon mode. The E_{2g} mode does not exhibit any softening at T_c (see Ref. 30) and therefore does not provide the superconducting electron pairing. Nevertheless, it is responsible for the T_c behavior with pressure as we will see later. It should be pointed out that baric experiments have been completed both for pressed powder and single crystals. No structural transitions were found in experiments^{3,29,31} up to the pressure 40 GPa.

The goal of the present paper is to calculate the pressure derivative dT_c/dp . We assume that the large polarons are formed in the boron planes due to the σ -electron interaction with the optical E_{2g} phonons.⁷ Because of the polaron for-

mation the σ band is lowered while the π electrons are not influenced. As the events take place in the layered medium we consider the quasi-two-dimensional polaron. The polaron anisotropy parameter and, correspondingly, the σ -band energy shift depend on the lattice constants influenced by the pressure. Roughly speaking, the Cooper pairs of the correlated σ electrons in the MgB_2 superconducting condensate are plunged into the pancake polaron. To treat the Cooper pairing we adopt the kinematic mechanism^{32,33} in MgB_2 .

II. FORMULATION OF THE MODEL

In order to describe the superconducting state of MgB_2 , we adopt the model with the strongly interacting σ electrons (with a narrow bandwidth $2w_1$) overlapping with the non-correlated π electrons (with a wide bandwidth $2w_2$) of boron ions. The important parameter here is the distance r between the centers of the σ and π bands influenced by the pressure.

The positive Hall current and the thermoelectric power measurements³⁴ indicate the hole conductivity in MgB_2 . The polaron radius is given by the expression

$$R \sim \sqrt{\frac{\hbar}{m\omega}} = \frac{87.3 \text{ \AA}}{\sqrt{(m/m_e)(\hbar\omega/1 \text{ meV})}}, \quad (1)$$

where m and m_e are the hole and the vacuum electron masses, respectively, and $\hbar\omega$ is the low-frequency optical phonon energy responsible for the polaron formation. Inserting the value $\hbar\omega \approx 74.5$ meV for the E_{2g} phonons and the light-hole band mass³⁵ $m = 0.25m_e$ into Eq. (1), the polaron radius can be estimated as $R \approx 20$ \AA, which is much larger than the lattice constant.

Studying the polaron sector of the system, we apply the conventional Pekar-Fröhlich Hamiltonian generalized for the consideration of the peculiarities of MgB_2 . To take into account the fact that the σ electrons are located in narrow layers inhabited by boron atoms we introduce the in-plane mass m , which is the light-hole band mass, and the large mass M in the orthogonal (z) direction. The ratio m/M of these masses is related to the probability of a carrier to tunnel from one layer to another.

To take into account the different polarizability of the medium in the xy plane and in the z direction we suggest to simulate it by using the anisotropic Coulomb potential for the interaction of the hole with the induced polarization field. That is, we use the potential of the form $V(\vec{r}) \propto 1/\sqrt{x^2 + y^2 + (z/\gamma)^2}$ with $\gamma \ll 1$ being a phenomenological parameter to describe the deformation of the Coulomb potential along the z axis (that is, the change to an oblong form of the equipotential surface). In the momentum space the potential is of the form $\tilde{V}(\vec{k}) \propto 1/(k_{\parallel}^2 + \gamma^2 k_z^2)$.

In a sense we study the electron sector since all events happen to be located inside the large polarons. With large polaron formed, we obtain a lowering of the energy of carriers, that is, the renormalization of the parameter r , which becomes dependent on the applied pressure. This influences drastically the electron structure of MgB_2 -like systems, especially the relative position of σ and π bands.

A. Polaron sector

Thus, we consider an anisotropic polaron whose motion is confined to the xy plane while the motion in the perpendicular z direction (along the crystallographic c axis of MgB_2) is restricted. The Pekar-Fröhlich Hamiltonian reads as follows:

$$H = \frac{\vec{p}_{\parallel}^2}{2m} + \frac{p_z^2}{2M} + \hbar\omega \sum_{\vec{k}} a_{\vec{k}}^{\dagger} a_{\vec{k}} + \frac{1}{\sqrt{\Omega}} \sum_{\vec{k}} (a_{\vec{k}} \tilde{V}_{\vec{k}} e^{i\vec{k} \cdot \vec{r}} + a_{\vec{k}}^{\dagger} \tilde{V}_{\vec{k}}^* e^{-i\vec{k} \cdot \vec{r}}), \quad (2)$$

where Ω is the crystal volume, \vec{p}_{\parallel} and p_z are the hole momenta in the xy plane and in the orthogonal z direction, respectively, and ω is the LO-phonon frequency. Operators $a_{\vec{k}}^{\dagger}$ ($a_{\vec{k}}$) are the creation (annihilation) operators of the phonons with momentum \vec{k} . The electron-phonon interaction term is specified by $\tilde{V}_{\vec{k}}$; its squared modulus is the Fourier transform of the Coulomb potential $V(\vec{r})$. In contrast with the conventional definition we introduce an additional anisotropy of the Coulomb potential to describe the different polarizability of the medium in various directions (in the isotropic boron plane and perpendicular to it):

$$\tilde{V}_{\vec{k}} = -i\hbar\omega \left(\frac{4\pi\alpha}{k_{\parallel}^2 + \gamma^2 k_z^2} \sqrt{\frac{\hbar}{2m\omega}} \right)^{1/2}. \quad (3)$$

Here α is the conventional Fröhlich coupling constant of the electron-phonon interaction.

The MgB_2 pressure compression is anisotropic.^{2,29,36} According to Ref. 29 the compressibility along the c axis is almost twice as large as the plane compressibility. Under hydrostatic pressure the initial compression along the c axis is larger than along the boron plane.³⁷ We estimate the parameter γ as the ratio of the standard lattice constants: $\gamma \sim c/2a$ (here $c/2$ is the distance between charged boron and magnesium planes). When the pressure increases, the distance between the Mg and the B planes decreases, which is described by a decrease of the Coulomb potential anisotropy γ in our model. The polaron self-energy ΔE can be found within the second order of the perturbation theory:

$$\Delta E = -\frac{\alpha\hbar\omega}{2\pi^2} \int \frac{d^3k}{(k_{\parallel}^2 + \gamma^2 k_z^2)[k_{\parallel}^2 + (m/M)k_z^2 + 1]}. \quad (4)$$

With evaluation of integration in Eq. (4), we arrive at the expression:

$$\Delta E = -\alpha\hbar\omega \sqrt{\frac{M}{m}} \times \begin{cases} \frac{1}{\sqrt{1-\Gamma^2}} \ln \frac{1+\sqrt{1-\Gamma^2}}{\Gamma}, & \Gamma \leq 1; \\ \frac{1}{\sqrt{\Gamma^2-1}} \arctan \sqrt{\Gamma^2-1}, & \Gamma \geq 1. \end{cases} \quad (5)$$

Here the parameter $\Gamma = \gamma\sqrt{M/m}$. This formula reproduces two well-known limiting cases. When $M = m$ and $\gamma = 1$, then

$\Gamma=1$ and we obtain $\Delta E = -\alpha\hbar\omega$, which is the conventional result for a three-dimensional polaron. When $\gamma=1$ and $M \gg m$, we arrive at the result $\Delta E = -(\pi/2)\alpha\hbar\omega$, which is valid for a two-dimensional polaron confined to a plane.^{38,39} If γ is finite and $M \gg m$, then $\Gamma \gg 1$ and Eq. (5) leads to the following expression:

$$\Delta E = -\frac{\pi}{2\gamma}\alpha\hbar\omega. \quad (6)$$

It is the MgB₂ case indeed for the reason that the mass ratio is inversely proportional to the hopping integral ratio: $M/m \propto t/t_z \propto 10$ (see Ref. 40). For the numerical estimates we will take the experimental value for the optical phonon frequency ω . The energy shift ΔE is negative, which means that the total lowering of the band minimum of the hole carriers is due to the polaron effect. Because of ΔE the energy distance r between the σ and π bands is shifted by a frequency-dependent contribution, which is essential for the electron sector.

B. Electron sector

We start from the high-temperature paramagnetic phase of the MgB₂ system and derive the superconducting critical temperature from the condition of instability of the normal state of correlated σ electrons due to a temperature decrease. Namely, T_c is governed by the solution of the Bethe-Salpeter equation for a vertex Γ_p in the Cooper channel in the reference frame of the electron pair:

$$\Gamma_p = -T \sum_{n,q} [-2t_q + V(p-q)] G_{\omega_n}^{0+}(q) G_{-\omega_n}^{0-}(-q) \Gamma_q, \quad (7)$$

where $G_{\omega_n}^{0s}(q) = 1/(-i\omega_n + \xi_q)$ is the normal state Green's function for a strongly correlated σ electron with a spin orientation s and an energy dispersion $\xi_q = ft_q - \mu$ with the correlation factor⁴¹

$$f = \frac{2w_1 + w_2 + 3r}{5w_1 + 4w_2}. \quad (8)$$

The Matsubara frequencies are given by $\omega_n = (2n+1)\pi T$ in Eq. (7).

The expression for the chemical potential

$$\mu = w_1 \frac{w_2 - 5r}{5w_1 + 4w_2} \quad (9)$$

follows from the equation $n_\sigma + n_\pi = 2$ for the total electron density per boron with the assumption of complete ionicity of Mg ions in the Mg²⁺[B⁻($p^{n_p} + n_\sigma$)]₂ system. In the integral kernel of Eq. (7) near the Γ -A line of the Brillouin zone, the Coulomb vertex $V(p-q)$ can be factorized as

$$V(p-q) = 2\beta t_p t_q, \quad (10)$$

where the parameter $\beta = V/6t^2$ labels an effect of the Coulomb repulsion for the nearest σ electrons, and the energy

dispersion is $t_p = 3t[1 - (p_x^2 + p_y^2)/12]$ near the Γ -A line. The kernel of the integral equation (7) does not contain other kinematic vertices⁴¹ that could be essential for superconducting condensates at moderate densities of carriers and/or for specific symmetries of superconducting order parameters.

Upon summation over the Matsubara frequencies, Eq. (7) takes the form

$$1 = \sum_q t_q \frac{1 - \beta t_q}{\xi_q} \tanh \frac{\xi_q}{2T_c}. \quad (11)$$

The superconducting coupling constant can be written as

$$\lambda = \frac{\mu}{w_1 f^2} \left(1 - \frac{\beta}{f} \mu \right), \quad (12)$$

so the chemical potential is restricted by the inequality $0 \leq \mu \leq f/\beta$, which generates constraints for the electron structure parameters $w_{1,2}$ and r . The larger the electron-electron Coulomb repulsion $V \sim \beta$ the narrower the superconducting region and the lower the T_c .

Hereafter we will neglect the interelectron Coulomb repulsion ($\beta=0$). Then the superconducting critical temperature satisfies the following equation:

$$1 = \int_{\xi(-w_1)}^{\xi(w_1)} d\xi \rho(\xi) \frac{\xi + \mu}{\xi f^2} \tanh \frac{\xi}{2T_c}, \quad (13)$$

where $\xi(w_1)$ and $\xi(-w_1)$ are the energy dispersion values of σ electrons at the top and the bottom of their energy band, respectively, and $\rho(\xi)$ is the electronic density of states. Equation (13) for the superconducting critical temperature coincides with that derived in Refs. 32 and 33 in a different way.

Note that the kinematic superconducting mechanism was also applied to MgB₂ in Ref. 42 but with a nonphysical negligence of the π -electron role. In our approach the great importance has the characteristic energy difference r_0 between the σ and π bands, shifted by ΔE due to the interaction between the light holes in the σ band and the quasi-two-dimensional E_{2g} phonons:

$$r = r_0 + \Delta E. \quad (14)$$

Due to the chemical bond nature, the hydrostatic pressure decreases the interplane distance more readily than the in-plane boron-boron distance, so that $dw_2/dp \gg dw_1/dp$ (cf. Ref. 43), and the latter derivative can be neglected in our estimates. The calculations of $T_c(r)$ demonstrate³² that $r \geq -w_1/4$ for MgB₂. As the next step we differentiate the integral equation (13) with respect to the pressure and take Eq. (14) into account. Assuming the rectangular density of states $\rho = \theta(w_1^2 - \xi^2)/2w_1$, we obtain after subsequent integration the following expression:

$$\frac{d \ln T_c}{dp} = \frac{1}{5w_1 + 4w_2} \left[\frac{2w_1}{T_c} - 1 - 5 \ln \left(\frac{\gamma_0 w_1}{\pi T_c} \right) \right] \frac{dr}{dp}, \quad (15)$$

where $\gamma_0 = \ln C$ with $C = 0.577$ being the Euler's constant. Under the natural assumption $T_c \ll w_1$ one can keep only the first term in the brackets in Eq. (15). Then we arrive at the result

$$\frac{dT_c}{dp} = \frac{2w_1}{5w_1 + 4w_2} \frac{d\Delta E}{dp}. \quad (16)$$

Taking into account that the Fröhlich coupling constant $\alpha \propto 1/\sqrt{\omega}$, we find then from Eq. (6):

$$\frac{d\Delta E}{dp} = \Delta E \frac{d \ln(\sqrt{\omega}/\gamma)}{dp}. \quad (17)$$

The parameter $\gamma = c/2a$ evidently decreases with pressure. As to the quasi-two-dimensional frequency ω we use the estimate of a quasiharmonic phonon via the mode Grüneisen parameter $d \ln \omega / dp = G/B_0$. The quantity B_0 is said to be the bulk modulus. It follows then that both ω and $\sqrt{\omega}/\gamma$ increase with the pressure. These findings are compatible with an experimental study^{29,44} of Raman spectra and lattice parameters of MgB₂ under pressure at room temperature and with theoretical estimations⁴⁵ of the strong influence of the pressure on the E_{2g} mode. Finally we conclude that $d\Delta E/dp < 0$ and $dT_c/dp < 0$. In an explicit form the baric derivative for the superconducting critical temperature can be written as follows:

$$\frac{dT_c}{dp} = - \frac{|\Delta E|}{5 + 4w_2/w_1} \left(- \frac{d \ln \gamma^2}{dp} + \frac{G}{B_0} \right). \quad (18)$$

III. NUMERICAL RESULTS AND CONCLUSIONS

For numerical calculations we estimate the polaron anisotropy parameter as $\gamma = c/2a = 0.57$. The known value of its derivative³⁷ is $d \ln \gamma^2 / dp = 2d \ln(c/2a) / dp \approx -2.4 \times 10^{-3} \text{ GPa}^{-1}$. The bulk modulus is measured⁴⁶ to be $B \approx 114 \text{ GPa}$. The Grüneisen parameter reported in Ref. 28 equals $G = 2.9 \pm 0.3$ for the measured Raman active E_{2g} -phonon mode with the energy $\hbar\omega = 76.7 \text{ meV}$, which we will use in Eq. (18). In particular, the polaron self-energy $\Delta E = -211.4\alpha \text{ meV}$. This polaronic shift could be useful to provide the proper Fermi surface areas for description de Haas-van Alphen data in MgB₂ (see Ref. 47). The realistic value for the MgB₂ energy band width ratio of π and σ electrons is $w_2/w_1 \approx 18/9 = 2$ (see Ref. 48).

Putting these magnitudes in Eq. (18), we obtain the estimate for the derivative of T_c with respect to pressure:

$$\frac{dT_c}{dp} \approx -\alpha(5.2 \pm 0.9) \frac{\text{K}}{\text{GPa}}. \quad (19)$$

The uncertainty of the result comes from the experimental deviation errors in the Grüneisen parameter. Comparison with the low-pressure result^{1,2} $dT_c/dp = -1.2 \text{ K/GPa}$ leads to the interval $\alpha = 0.20 - 0.28$ for the Fröhlich electron-phonon coupling constant. Comparison with the result^{1,3,4} -2 K/GPa leads subsequently to the estimate $\alpha = 0.33 - 0.47$.

These results are shown in Fig. 1. The solid line presents the dependence of the pressure derivative dT_c/dp on the

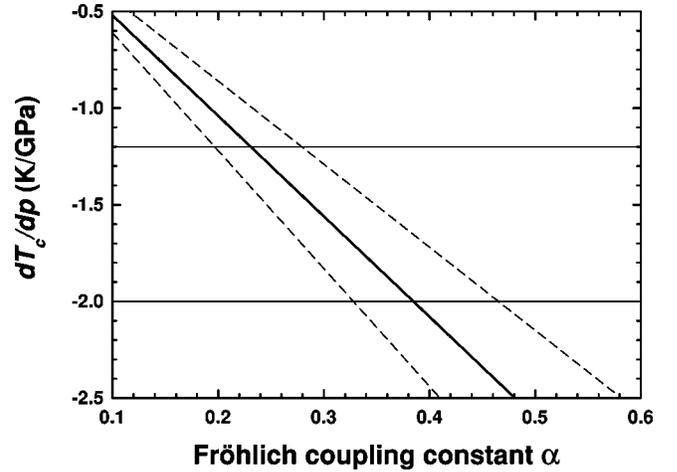


FIG. 1. Dependence of the pressure derivative of the superconducting critical temperature on the Fröhlich electron-phonon coupling constant (solid line). The dashed lines show error bars for α due to the uncertainty in the Grüneisen parameter (Ref. 29). The thin solid lines show experimental values (Refs. 1–4) for dT_c/dp .

electron-phonon coupling constant α . The thin solid lines represent the experimentally measured values for dT_c/dp and the dashed lines show error bars for α at a given value of the pressure derivative.

We note that the numerical results can be changed if one scales the frequency shift of the in-plane mode with the variation of the interatomic bond distance or lattice parameter.⁴⁹ Then the Grüneisen parameter takes even the larger value²⁹ $G = 3.9 \pm 0.4$ and our Eq. (19) takes the form:

$$\frac{dT_c}{dp} \approx -\alpha(6.9 \pm 1.1) \frac{\text{K}}{\text{GPa}}. \quad (20)$$

Subsequently, the estimates for the electron-phonon coupling constant will be different as well: $\alpha = 0.15 - 0.21$ for $dT_c/dp = -1.2 \text{ K/GPa}$ and $\alpha = 0.25 - 0.34$ for $dT_c/dp = -2 \text{ K/GPa}$.

Combining the numerical results for α , we may conclude that the electron-phonon coupling constant in MgB₂ is in the range $\alpha = 0.15 - 0.45$, which seems to be a quite reasonable interval for the assumed weak-coupling regime of a large hole polaron.

We have proposed a model in which large anisotropic polarons play an important role in decreasing the energy distance r between the σ and π bands. The polaron anisotropy is governed by the introduced parameter γ , which depends on the geometry of the MgB₂ system influenced by pressure. In addition, the phonon frequency increases with the pressure, which also results in a decrease of the polaron radius. Thus, the superconducting properties of the MgB₂ system are influenced both by the geometry of the crystal and by the polaron well depth and size.

The superconducting instability is driven by the non-phonon kinematic mechanism with T_c depending on the energy difference r between the σ and π bands. The quantity r incorporates all electron-phonon effects in our model. As the result the pressure derivative dT_c/dp is calculated as a func-

tion of the Fröhlich coupling constant α . At realistic values of α our calculations agree with experiments. It follows from the arguments discussed in the present paper that the pressure measurements provide us with valuable tests to establish the acceptable model for MgB_2 , to understand the mechanism of the superconductivity in this material, and to estimate its Fröhlich electron-phonon coupling constant.

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