## Pressure effect on the magnetic penetration depth in MgB<sub>2</sub>

D. Di Castro, <sup>1,2,\*</sup> R. Khasanov, <sup>1,3,4</sup> C. Grimaldi, <sup>4,5</sup> J. Karpinski, <sup>6</sup> S. M. Kazakov, <sup>6</sup> R. Brütsch, <sup>7</sup> and H. Keller <sup>1</sup> Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland <sup>2</sup> INFM-Coherentia and Dipartimento di Fisica, Universita' di Roma "La Sapienza," P.le A. Moro 2, I-00185 Roma, Italy <sup>3</sup> Laboratory for Neutron Scattering, ETH Zürich and Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland <sup>4</sup>DPMC, Université de Genève, 24 Quai Ernest-Ansermet, 1211 Genève 4, Switzerland <sup>5</sup> Ecole Polytechnique Fédérale de Lausanne, LPM, CH-1015 Lausanne, Switzerland <sup>6</sup> Solid State Physics Laboratory, ETH, CH-8093 Zürich, Switzerland <sup>7</sup> Laboratory for Material Behaviour, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland (Received 28 November 2005; revised manuscript received 1 June 2005; published 13 September 2005)

A study of the pressure effect on the magnetic penetration depth  $\lambda$  in polycrystalline MgB $_2$  was performed by measuring the temperature dependence of the magnetization under an applied pressure of 0.15 and 1.13 GPa. We found that  $\lambda^{-2}$  at low temperature is only slightly affected by pressure  $[\Delta \lambda^{-2}/\lambda^{-2}=3.1(9)\%]$ , in contrast to cuprate superconductors, where, in the same range of pressure, a very large effect on  $\lambda^{-2}$  was found. Theoretical estimates indicate that most of the pressure effect on  $\lambda^{-2}$  in MgB $_2$  arises from the electron-phonon interaction.

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Shortly after the discovery of the superconductivity in MgB<sub>2</sub> at 39 K, several investigations of the pressure dependence of the superconducting critical temperature  $T_c$  were carried out.<sup>1-4</sup> Indeed, the magnitude and sign of  $dT_c/dp$ may indicate a way to raise  $T_c$  at ambient pressure, and moreover may help to obtain information about superconducting pairing mechanism. So far, all these studies show that  $T_c$  decreases with increasing pressure, with a rate depending on the method and the pressure medium used. The first hydrostatic measurement of  $T_c(p)$  up to 0.7 GPa, reveals that  $T_c$  decreases reversibly under hydrostatic pressure at the rate  $dT_c/dp = -1.11(2)$  K/GPa.<sup>4</sup> The behavior of  $T_c(p)$  with pressure in MgB<sub>2</sub> was attributed to the pressure induced lattice stiffening (increase of the phonon frequency), 5,6 rather than to the decrease in the electronic density of states  $N(E_F)$ , that is only moderately affected by pressure.<sup>7</sup> Comparison with theoretical calculations supported the view that MgB<sub>2</sub> is a BCS superconductor with moderately strong electronphonon coupling.5

Apart from  $T_c$ , another relevant superconducting parameter is the magnetic field penetration depth  $\lambda$ . In fact the so-called superfluid density  $\lambda^{-2}$  is related to the Fermi velocity and to the density of charge carriers, and its temperature dependence gives information on the symmetry and on the magnitude of the superconducting gap. A study of pressure effects on  $\lambda^{-2}(0)$  can give important information on how the electronic degrees of freedom are affected by lattice modifications and on the nature of the electron-phonon coupling. Indeed, in cuprates high temperature superconductors (HTS), a huge pressure effect on  $\lambda^{-2}(0)$  was found, in particular in YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub> (Y124).<sup>8</sup> Part of this effect was attributed to the strong renormalization of the Fermi velocity, and therefore to the effective mass, due to the nonadiabatic coupling of the electrons to the lattice. This result is consistent with the substantial oxygen isotope effect found in cuprates<sup>9–13</sup> and can be interpreted in the framework of nonadiabatic theory of the electron-phonon interaction. Indeed, the failure of the Migdal (adiabatic) approximation, occurring when the typical phonon frequency  $\omega_{ph}$  is comparable to the Fermi energy  $E_F$ , has been shown to open interaction channels giving rise to, e.g., anomalous pressure and isotope effects. <sup>14–16</sup> Anomalous features are expected to characterize also the polaronic regime. <sup>17</sup>

Although it was proposed<sup>18</sup> that also MgB<sub>2</sub> is a nonadiabatic superconductor, because of the large coupling strength of the electrons to the E<sub>2g</sub> phonon mode and the small value of  $E_F$  of the  $\sigma$  bands relative to the phonon energy  $\omega_{ph}$  (which could violate the adiabatic assumption  $\omega_{ph} \ll E_F$ ), the negligible boron isotope effect on  $\lambda(0)$  found in MgB<sub>2</sub> (Ref. 19) suggested that in this system the nonadiabatic effects on  $\lambda(0)$  are not relevant. However, we note that the proximity of the Fermi level to the top of the  $\sigma$  bands may result in an apparent adiabaticity.<sup>16</sup>

In this paper we report measurements of the magnetic penetration depth under pressure in polycrystalline MgB<sub>2</sub> to gain additional information on the nature of the electron-phonon coupling, and to check if, as in cuprates, we obtain results consistent with those obtained from the boron isotope effect measurements.<sup>19</sup> The temperature dependence of  $\lambda^{-2}(T)$  was extracted from low field magnetization measurement. For pressure ranging from 0.15 to 1.13 GPa a small pressure (p) effect on  $\lambda^{-2}(0)$  was found  $[\Delta\lambda^{-2}/\lambda^{-2}=3.1(9)\%]$ , compared to the huge effect  $(\sim40\%)$  on  $\lambda^{-2}(0)$  found in the nonadiabatic cuprate superconductor Y124 in the same range of pressure.<sup>8</sup> We report also theoretical calculations of the pressure effect on  $\lambda(0)$ , which confirm the smallness of this effect.

The  $MgB_2$  powder sample was prepared by solid state reaction in flowing argon. As starting materials we used Mg flakes and amorphous boron (Alfa Aesar). A pellet with starting composition  $Mg_{1.1}B_2$  was placed in a BN crucible and fired in a tube furnace under pure Ar gas. The sample was heated for one hour at  $600~^{\circ}C$ , one hour at  $800~^{\circ}C$ , and one hour at  $900~^{\circ}C$ .

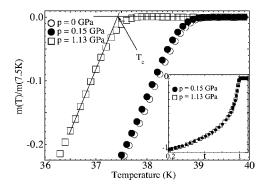


FIG. 1. Field cooled (0.5 mT) normalized magnetization of MgB<sub>2</sub> as a function of temperature in the vicinity of  $T_c$  for  $p_Z$  (open circles),  $p_L$  (filled circles), and  $p_H$  (open squares). The inset shows the full normalized magnetization data for  $p_L$  and  $p_H$  as a function of the reduced temperature  $t=T/T_c$ . Note that some of the data points were dropped for clarity.

The sample was first ground and then sieved in order to obtain a small grain size, needed for the determination of  $\lambda$  from magnetization measurements. The grain size distribution was measured and shown in the inset of Fig. 2. The hydrostatic pressure was produced in a copper-beryllium piston cylinder clamp, especially designed for magnetization measurements under pressure (see Ref. 20). The sample was put in a Teflon cylinder and the pressure cell was then filled with fluorinert FC77 as pressure transmitting medium. The sample-to-liquid ratio in the pressure cell is about 1/8, small enough to consider each grain isolated by the other. The low field magnetization measurements (field cooling) were performed with a commercial superconducting quantum interference device.

For fine powders with known grain sizes the magnetic penetration depth can be calculated from the measured magnetization by using the Shoenberg formula,<sup>21</sup> modified for the known grain-size distribution  $N(R)^{22}$ 

$$\chi(T) = -\frac{3}{2} \int_0^\infty \left( 1 - \frac{3\lambda(T)}{R} \coth \frac{R}{\lambda(T)} + \frac{3\lambda^2(T)}{R^2} \right) g(R) dR /$$

$$\int_0^\infty g(R)dR,\tag{1}$$

where R is the grain radius and  $g(R) = N(R)R^3$ . Any change in  $\chi(T)$  due to pressure can be attributed mainly to a change of  $\lambda(T)$ , and much less to a change of R. The reducing of R with pressure is taken into account in calculating  $\lambda(T)$ , by using the bulk modulus reported in Ref. 6. The sample was measured at low pressure  $p_L = 0.15$  GPa and at the highest pressure available  $p_H = 1.13$  GPa. The pressure values were determined from a preliminary calibration measurement, where part of the same sample was measured in the pressure cell together with a small piece of lead. The pressure was detected by measuring the  $T_c$  shift of lead  $(T_c(0 \text{ GPa}) = 7.2 \text{ K}).^{24}$  We found  $dT_c/dp = -1.24(5) \text{ K/GPa}$ , in good agreement with previous results. The real zero-pressure measurement could not be performed in the pressure cell,

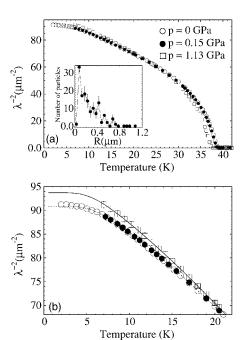


FIG. 2. (a)  $\lambda^{-2}$  as a function of temperature for  $p_Z$ =0 GPa (open circles),  $p_L$ =0.15 GPa (filled circles), and  $p_H$ =1.13 GPa (open squares). (b) Low temperature region on a larger scale. The error bars for the  $p_Z$  data are not shown for clarity. The dashed and solid lines are fits to the  $p_L$  and  $p_H$  data, respectively, using Eq. (4). Note that the data points are much more dense than what is shown in this figure, since part of them were dropped for clarity.

since, in order to seal the cell, at least a small pressure had to be applied.

In Fig. 1 the temperature dependence of the normalized magnetization of MgB<sub>2</sub> at  $p_L$  and  $p_H$  is shown in the vicinity of  $T_c$ , together with the zero pressure  $p_Z$  measurement performed on the same sample in a quartz tube. The  $T_c$ 's were obtained from the intercept of the linear extrapolations (see Fig. 1):  $T_c(p_Z)$ =38.73(3) K,  $T_c(p_L)$ =38.66(3) K, and  $T_c(p_H)$ =37.43(3) K. The magnetization curves shift systematically with increasing pressure toward lower temperature. In the inset of Fig. 1 we show the normalized magnetization as a function of the reduced temperature t= $T/T_c$  for  $p_L$  and  $p_H$ . The identical temperature dependences for both pressure values indicate the absence of stresses in the sample due to pressure.

From the magnetization the susceptibility was calculated. Because of the unknown mass of the sample in the pressure cell, the susceptibility for the lowest pressure ( $p_L$  =0.15 GPa) was normalized at low temperature with the value obtained by measuring the same sample in a quartz tube at zero pressure  $p_Z$ . To calculate the susceptibility for the high pressure data, we took into account the volume contraction of the sample due to the pressure, by using the bulk modulus estimated in Ref. 6. The values of  $\lambda^{-2}$  were then calculated using the modified Shoenberg formula [Eq. (1)].

We define the shift of  $\lambda^{-2}$  between two different pressures  $p_L$  (lower) and  $p_H$  (higher) at a temperature T as

$$\frac{\Delta \lambda^{-2}}{\lambda^{-2}} \equiv \frac{\lambda^{-2}(p_H, T) - \lambda^{-2}(p_L, T)}{\lambda^{-2}(p_L, T)}.$$
 (2)

	<i>T</i> (K)	$p_L$ (GPa)	$p_H$ (GPa)	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
Exper.	≃7	0.15	1.13	2.8(1.1)
Fit	0	0.15	1.13	3.1(9)
Theory	0	0.15	1.13	$\approx 1.4 \div 2.2$

TABLE I. Summary of the experimental and theoretical estimations of the pressure shift of  $\lambda^{-2}$ .

The temperature dependences of  $\lambda^{-2}$  for  $p_L$  and  $p_H$ , are shown in Fig. 2, together with the  $p_7$  data measured in the quartz tube. Unfortunately, we were not able to obtain reliable data below 7 K because of a background signal due to lead impurities in the composition of the pressure cell. This background signal is negligible above 7 K. This is demonstrated by the fact that the  $\lambda^{-2}(T)$  curves for  $p_Z$  and  $p_L$  overlap perfectly for temperatures not too close to  $T_c$ , where a small pressure shift is present. Therefore, while for the  $p_Z$ measurement in the quartz tube we show the data down to 2 K, for the  $p_L$  and the  $p_H$  measurements we show the data only down to  $\approx$ 7 K (see lower panel of Fig. 2). Due to the pressure effect on  $T_c$ , the curves for  $p_L$  and  $p_H$  are clearly distinct at high temperature, but they tend to merge at low temperature. The error bars on  $\lambda^{-2}$  [shown in Fig. 2(b)] were determined considering that magnetization measurements are reproducible within  $\approx 0.5\%$ . The value of  $\lambda^{-2}$  for the  $p_7$  measurement at the lowest temperature (2.5 K) is in good agreement with that one measured with the muon spin rotation technique on the same sample at zero pressure.<sup>25</sup>

The shift of  $\lambda^{-2}$  at the lowest temperature available  $T \approx 7$  K between  $p_L = 0.15$  GPa and  $p_H = 1.13$  GPa [see Eq. (2)] is

$$\Delta \lambda^{-2} / \lambda^{-2} = 2.8(1.1)\%.$$
 (3)

This result shows that a small pressure effect on  $\lambda^{-2}$  is present. The pressure results are summarized in Table I.

To obtain an estimation of the pressure effect on the zero temperature  $\lambda^{-2}(0)$ , a fit to the experimental data is needed. It was well established that MgB2 is a two band superconductor with two superconducting gaps of different size, the larger one originating from the two-dimensional (2D)  $\sigma$  band and the smaller one from the 3D  $\pi$  band.  $^{26-29,32}$  Taking this into account, the temperature dependence of  $\lambda$ , in the low temperature range, can be written in the form  $^{30}$ 

$$\lambda^{-2}(T) = \lambda^{-2}(0) \left[ 1 - w \left( \frac{2\pi\Delta_1(0)}{k_B T} \right)^{1/2} \exp\left( -\frac{\Delta_1(0)}{k_B T} \right) - (1 - w) \left( \frac{2\pi\Delta_2(0)}{k_B T} \right)^{1/2} \exp\left( -\frac{\Delta_2(0)}{k_B T} \right) \right]. \tag{4}$$

Here,  $\Delta_1$  and  $\Delta_2$  are the zero temperature small and large gap associated to the  $\pi$  and  $\sigma$  bands, respectively, and  $w = \lambda_1^{-2}(0)/\lambda^{-2}(0)$ .

The zero pressure data were then fitted to Eq. (4) up to 22 K. The fit gave the following results:  $\lambda^{-2}(0) = 91.1(2)~\mu\text{m}^{-2},~w=0.23(3);~\Delta_1=2.3(2)~\text{meV},~\text{and}~\Delta_2=7.2(4)~\text{meV}.$  The estimated gaps are in good agreement with previous results from penetration depth measurements.  $^{30,31}$ 

Because of the lack of data below 7 K for  $p_L$  and  $p_H$ , these data were analyzed with fixed w obtained from the fit to the zero-pressure data. Here it is assumed that w is not affected by pressure up to 1.13 GPa. This assumption will be shown below to be correct by our model calculations [see Eq. (9)]. The other fitting parameters  $[\lambda^{-2}(0)]$  and the gaps were left free. As shown by the solid and dashed lines in panel (b) of Fig. 2, the data are well described by Eq. (4). In particular the fitted curve of the  $p_L$  data (dashed line) follows very well the  $p_Z$  data below 7 K. This indicates that, although the data for the sample under pressure are limited down to 7 K, that is, above the region where  $\lambda^{-2}(T)$  becomes flat, however it is highly reliable that the fitting curve well reproduces the missing data below this temperature. The fit reproduces the missing data colonial.  $\mu_L = 90.9(5) \mu^{-2}$  and  $\lambda^{-2}(0)_{p_L} = 90.9(5) \mu^{-2}$  and  $\lambda^{-2}(0)_{p_L} = 90.9(5) \mu^{-2}$ =93.7(6)  $\mu$ m<sup>-2</sup> (see Table II). So that the relative shift of  $\lambda^{-1}$ at T=0 between  $p_L=0.15$  GPa and  $p_H=1.13$  GPa is [see Eq. (2)

$$\frac{\Delta \lambda^{-2}}{\lambda^{-2}} = 3.1(9)\%,\tag{5}$$

a value in agreement with the experimental one obtained directly form the data at 7 K [Eq. (3)]. This result indicates

TABLE II. Fitting parameters obtained from the fit of the experimental data shown in Fig. 2 by Eq. (4).

p (GPa)	$\lambda^{-2}(0) \ (\mu \text{m}^{-2})$	$\begin{array}{c} \Delta_1(0) \\ (\text{meV}) \end{array}$	$\begin{array}{c} \Delta_2(0) \\ (\text{meV}) \end{array}$	w	$2\Delta_1(0)/k_BT_c$	$2\Delta_2(0)/k_BT_c$
0	91.1(2)	2.3(2)	7.2(4)	0.23(3)	1.38(12)	4.31(24)
0.15	90.9(5)	2.4(1)	7.1(1)		1.44(6)	4.26(6)
1.13	93.7(6)	2.3(1)	6.8(1)		1.43(6)	4.28(6)

that there is a small, but not zero, pressure effect on the magnetic penetration depth in the range p=0.15–1.13 GPa. Although not zero, this effect found in the MgB<sub>2</sub> system is much smaller than that one found in the Y124 cuprate superconductor, confirming the different character of electron-phonon coupling in these two systems. A summary of the pressure results are reported in Table I. All the fitting parameters are summarized in Table II. The two gaps show a small decrease with increasing pressure, in agreement with the corresponding decrease of  $T_c$ . In Table II we added also the ratio  $2\Delta/k_BT_c$  calculated for  $\Delta_1$  and  $\Delta_2$  at each pressure. This ratio appears to be pressure independent within errors.

Now let us turn to discuss how the experimental result (5) may be interpreted in terms of a theoretical model of MgB<sub>2</sub>. Considering that  $p_H - p_L \approx 1$  GPa, Eq. (5) indicates that the logarithmic derivative of  $\lambda^{-2}(0)$ ,  $d \ln \lambda^{-2}(0)/dp$ , must be positive and of the order of few %/GPa. Since  $\lambda^{-2}(0) \propto \omega_p^2$ , where  $\omega_p$  is the plasma frequency, then a free electron gas estimate would give only  $d \ln \lambda^{-2}(0)/dp = 1/B \approx 0.65\%$ /GPa, where  $B = -dp/d \ln \Omega \approx 155$  GPa is the bulk modulus of MgB<sub>2</sub>,<sup>6</sup> and  $\Omega$  the volume of the unit cell. Although being positive, the free electron estimate is however about five times smaller than that measured. Let us try then to improve the calculations by considering the band structure effects of MgB<sub>2</sub>. To this end, let us consider the zero temperature expression of the penetration depth<sup>33</sup>

$$\lambda^{-2}(0) = \frac{e^2}{3\hbar \pi^2 c^2} \sum_{n} \oint_{S_E^n} ds |\mathbf{v}_n(s)|,$$
 (6)

where the integral runs over the Fermi surface  $S_n^F$  of the nth band  $(n=\sigma,\pi)$  and  $\mathbf{v}_n(s)$  is the corresponding surface bare electron velocity vector. In the following, we use the model of Ref. 34 where the  $\pi$  bands are modeled by a half-torus Fermi surface of area  $S_1$  and Fermi velocity  $v_1$ , while the  $\sigma$  bands are approximated by a cylindrical Fermi surface of area  $S_2$  and Fermi velocity  $v_2$ . Equation (6) then reduces simply to  $\lambda^{-2}(0) = \lambda_1^{-2}(0) + \lambda_2^{-2}(0)$ , where  $\lambda_i^{-2}(0) \propto S_i v_i$ , i=1, 2, and

$$\frac{d\ln\lambda^{-2}(0)}{dp} = w\frac{d\ln\lambda_1^{-2}(0)}{dp} + (1-w)\frac{d\ln\lambda_2^{-2}(0)}{dp},$$
 (7)

where  $w = \lambda_1^{-2}(0)/\lambda^{-2}(0) = S_1 v_1/(S_1 v_1 + S_2 v_2)$  is the parameter introduced in Eq. (4). Within the same approximations, the electron density of states at the Fermi level reads  $N_F = N_1 + N_2$ , where  $N_i \propto \Omega S_i/v_i$  is the partial density of states for the band i = 1, 2. Therefore, since  $\lambda_i^{-2}(0) \propto S_i v_i \propto \Omega S_i^2/N_i$ , and considering that  $S_1$  and  $S_2$  scale as  $\Omega^{-2/3}$ , Eq. (7) reduces to

$$\frac{d \ln \lambda^{-2}(0)}{dp} = \frac{1}{3B} - \left[ \frac{1 - w + \eta w}{\eta + (1 - \eta)N_2/N_F} \right] \frac{d \ln N_F}{dp}, \quad (8)$$

where we have introduced the parameter  $\eta = (d \ln N_1/dp)/(d \ln N_2/dp)$  whose calculated value ranges between  $\eta \approx 1$ ,<sup>35</sup> and  $\eta \approx 0$ .<sup>36</sup> Hence, by setting  $d \ln N_F/dp \approx -0.31\%$  /GPa,<sup>7</sup> w = 0.22, and  $N_2/N_F \approx 0.4$ ,<sup>37</sup> from Eq. (8)

we obtain  $d \ln \lambda^{-2}(0)/dp \approx 0.8\%$  /GPa or 0.5%/GPa according to whether  $\eta$ =0 or  $\eta$ =1, respectively. By the same token, it can be shown that

$$\frac{dw}{dp} = \frac{w(1-w)(1-\eta)}{\eta + (1-\eta)N_2/N_F} \frac{d\ln N_F}{dp} \simeq (0-0.1) \% / \text{GPa}, (9)$$

confirming that the assumption dw/dp=0 used in obtaining Eq. (5) is sufficiently accurate.

The fact that our estimate  $\Delta \lambda^{-2}/\lambda^{-2} \approx (0.5-0.8)\%$  does not deviate from the free electron gas result suggests that factors other than band structure should be considered in order to get closer to the measured value reported in Eq. (5). It is then natural to consider the electron-phonon interaction  $\lambda_{el-ph}$ , whose strong pressure dependence in MgB<sub>2</sub> is basically given by a hardening of the optical phonon mode  $E_{2g}$ . The electron-phonon interaction affects  $\lambda^{-2}(0)$  through the renormalization of the Fermi velocity appearing in Eq. (6). Hence the electron-phonon renormalized penetration depth reduces to  $\lambda^{*-2}(0) = \lambda^{-2}(0)/(1+\lambda_{el-ph})^{38}$  where  $\lambda^{-2}(0)$  is the bare quantity we have considered before, and therefore

$$\frac{d \ln \lambda^{*-2}(0)}{dp} \simeq (0.5 - 0.8) \% / \text{GPa} - \frac{\lambda_{e-ph}}{1 + \lambda_{e-ph}} \frac{d \ln \lambda_{e-ph}}{dp}.$$
(10)

Different estimates of  $d \ln \lambda_{e\text{-}ph}/dp$  exist in literature<sup>7,36,39</sup> and range from  $\approx$ -1.7%/GPa,<sup>7</sup> up to about -2.8%/GPa.<sup>36</sup> Hence, by using  $\lambda_{el\text{-}ph}\approx$ 1,<sup>36,37</sup> we find  $\Delta\lambda^{*-2}/\lambda^{*-2}\approx$ (1.4-2.2)%, in better agreement with the measured value [Eq. (5)] (see also Table I for a direct comparison). This analysis evidences therefore that, although being rather small, the measured pressure effect on  $\lambda^{-2}(0)$  can be reasonably well understood only by considering the electron-phonon renormalization contribution.

Similar experiments on the Y124, in the same range of pressure, give a very large effect on  $\lambda(0)^{-2}(\sim 40\%).^8$  Arguments are used there to deduce that part  $(\sim 30\%)$  of this large effect is due to the pressure dependence of the Fermi velocity, because of the nonadiabatic electron-lattice coupling. From the results of the present work, we can therefore argue that such nonadiabatic effects on  $\lambda(0)^{-2}$  are negligible in MgB<sub>2</sub>, as previously suggested by  $\mu$ SR measurements. It should be however noted that the proximity of the Fermi level to the top of the  $\sigma$  bands in MgB<sub>2</sub> could mask some nonadiabatic features. If

In summary, we studied the pressure effect on the magnetic penetration depth at low temperature in polycrystalline MgB<sub>2</sub>. We found that pressure up to 1.13 GPa induces a small positive change in  $\lambda(0)^{-2} \left[\Delta \lambda^{-2}/\lambda^{-2} = 3.1(9)\%\right]$ , which is suggested to be due mostly to a pressure change of the electron-phonon coupling.

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- \*Email address: dicastro@physik.unizh.ch
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