Two-Band Effect on the Superconducting Fluctuating Diamagnetism in MgB2

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The field dependence of the magnetization above the transition temperature T_c in MgB₂ is shown to evidence a diamagnetic contribution consistent with superconducting fluctuations reflecting both the σ and π bands. In particular, the upturn field H_{up} in the magnetization curve, related to the incipient effect of the magnetic field in quenching the fluctuating pairs, displays a double structure, in correspondence with two correlation lengths. The experimental findings are satisfactorily described by the extension to the diamagnetism of a recent theory for paraconductivity, in the framework of a zero-dimensional model for the fluctuating superconducting droplets above T_c .

On approaching the transition temperature T_c from above, superconducting fluctuations (SFs) occur and, while the average order parameter is zero, one has $\langle |\psi|^2 \rangle^{1/2} \neq 0$. Therefore local pairs are generated, lacking long-range coherence and decaying with a characteristic time which increases for $T \rightarrow T_c^+$. An interesting way to detect the SF is by means of the related magnetic screening, through a diamagnetic contribution to the magnetization. In its simplest form this contribution is written $-M_{dia}(H,T) \approx$ $\frac{e^2}{mc^2}n_c \xi^2(T)H$, $n_c = |\psi|^2$ being the number density of superconducting pairs and $\xi(T)$ the coherence length. On cooling towards T_c^+ , because of the divergence of $\xi(T)$, $|M_{\text{dia}}|$ can be expected to enhance. On the other hand, strong magnetic fields, comparable to the critical field H_{c2} , must evidently suppress the SF. Thus the isothermal magnetization curves $-M_{\text{dia}}(H, T = \text{const})$ exhibit an upturn in the field dependence. According to a model of fluctuating superconductive particles of size $d \ll \xi(T)$ and located outside the critical region, the upturn field H_{up} is about inversely proportional to the square of $\xi(T)$ [1,2].

In conventional metallic BCS superconductors the diamagnetism above T_c and the effect of the field in quenching the fluctuating pairs were detected long ago, by means of magnetization measurements as a function of temperature at constant fields [3]. More recently, successful detection of the detailed field dependence of M_{dia} has been obtained in the new superconductor MgB_2 [4].

In Fig. 3 of the paper by Lascialfari *et al.* [4] an anomaly was noticeable: the curve of the reduced magnetization at $T = 39.5$ K (while $T_c \approx 39.05$ K) showed an unexplained double structure. Later on, in view of the established twoband character of the superconductivity in MgB_2 [5], it was suspected that the double structure in the magnetization curve could be related to the two superconducting bands in this compound. The motivation of the present Letter is mainly related to this hypothesis. In the following, we will show, for the first time, that also the fluctuation spec-

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trum above T_c reflects the presence of both the σ and π bands, from which variety of effects are known to occur in MgB_2 below T_c .

In this work, new magnetization measurements on a high-purity sample prepared by Palenzona *et al.* (University of Genova) have been carried out above $T_c = 39.1 \text{ K} \pm$ 0*:*04 K with high field resolution to evidence the double structure of the magnetization curves and to test its temperature behavior. The diamagnetic contribution was obtained by subtracting the paramagnetic contribution measured at 40 K (where the SF are practically ineffective) from the row data. The data obtained in a powdered sample at three representative temperatures are reported in Fig. 1.

FIG. 1. Diamagnetic contribution M_{dia} to the magnetization in $MgB₂$ as a function of the magnetic field *H* (after zero field cooling), for three temperatures above $T_c = 39.1 \pm 0.04$ K. No difference was observed for the magnetization in field-cooled condition at the same temperature (data not reported). In the inset the data for M_{dia} at a temperature close to T_c are reported, showing that for *H* smaller than the upturn field H_{up} the GL law showing that for *H* smaller than the upturn field H_{up} the GL law
in finite field, namely, $M_{\text{dia}}(H, T \approx T_c) \propto -\sqrt{H}$ (solid line), is well obeyed.

The field dependence of fluctuation dissipation can be understood in the framework of Ginzburg-Landau (GL) theory in a simple way, by resorting to a description based on fluctuation-induced superconducting droplets of spherical shape, with a diameter of the order of the coherence length. For these droplets, the so called zero-dimensional (0D) approximation can be used by assuming an order parameter is no longer spatial dependent. Then an exact solution for the GL functional is found in closed form, valid above the critical region and for all field $H \ll H_{c2}$. This was basically the framework used in the discussion of the data in Ref. [4].

Recently, SF in the presence of two superconducting bands has been studied in regards to paraconductivity and the specific heat in MgB_2 [6]. In this work a breakdown of the GL theory is hypothesized due to two different coherence lengths for the σ and π bands. In particular, the difference is relevant in the *z* direction, i.e., $\xi_{\sigma z} \ll \xi_{\pi z}$. An effective coherence length $\tilde{\xi}_z(T)$ is introduced, with $\xi_{\sigma z} \ll \tilde{\xi}_z(T) \ll \xi_{\pi z}$, in a large temperature range where a generalized nonlocalized GL model can be used [6].

A system with weakly coupled two bands labeled by 1 (in MgB₂ the σ band) and 2 (in MgB₂ the π band) is considered, and pairs can be formed only by electrons belonging to the same band. The pairing correlation matrix contains nondiagonal terms, allowing the pair to transfer from one band to the other. Dealing with the impurity scattering, it is known that in $MgB₂$ the interband contribution is very weak because of the different parity of the bands. Thus the transfer process can be neglected, and only the scattering frequency ν_1 and ν_2 in the two bands is considered. The inverse of the effective coupling matrix is introduced,

$$
\hat{W} = \begin{pmatrix} W_{11} & W_{12} \\ W_{21} & W_{22} \end{pmatrix},
$$

such that $W_{11}W_{22} - W_{12}W_{21} = 0$.

The free energy above T_c is determined by the fluctuation propagator [6]

$$
F = -k_B T V \int \frac{d^3 q}{(2\pi)^3} \ln \frac{A}{\det H_{\alpha\beta}},
$$
 (1)

where $H_{\alpha\beta}(q)$ is the linearized GL Hamiltonian density

$$
H_{\alpha\beta} = \begin{pmatrix} \nu_1(\xi_{1,\alpha}^2 q_\alpha^2 + W_{11} + \epsilon) & -\nu_2 W_{12} \\ -\nu_1 W_{21} & \nu_2 [W_{22} + \epsilon + f(\xi_{2z}^2 q_z^2) \xi_{2x}^2 q_\parallel^2 + g(\frac{2}{\pi^2} \xi_{2z}^2 q_z^2)] \end{pmatrix},
$$
(2)

with $\varepsilon = \frac{T-T_c}{T_c}$. Here $g(x) = \psi(1/2 + x) - \psi(x)$ and $f(x) = \frac{2}{\pi^2} g'(\hat{x})$, while $\xi_{1,a}^2 q_a^2 = \xi_{1x}^2 q_x^2 + \xi_{1y}^2 q_y^2 + \xi_{1z}^2 q_z^2$.

At a temperature close to T_c , where $\tilde{\xi}_z(T) \gg \xi_{2z}(0)$, only the long wavelength fluctuations of the order parameter can be taken into account. In this case, in the 0D approximation and when written in terms of the effective coherence length $\tilde{\xi}(T)$, the magnetization takes the form [2]

$$
M_{\text{dia}}(\varepsilon, H) = -K_B T H \frac{2\pi^2 \tilde{\xi}^2 d^2 / 5\Phi_0^2}{(\varepsilon + 2\pi^2 \tilde{\xi}^2 H^2 d^2 / 5\Phi_0^2)}.
$$
 (3)

Here Φ_0 is the flux quantum and *d* is the size of fluctuating droplets, which can be assumed to be of the order of $\tilde{\xi}(T)$. Equation (3) predicts a single upturn in the field dependence of M_{dia} , at a field $H_{\text{up}} \simeq \sqrt{5} \epsilon \Phi_0 / \pi \tilde{\xi}^2$.

For temperatures not too close to T_c , the short wavelength fluctuations are accounted for by a large value of the argument in the *g* and *f* functions in Eq. (2). In order to obtain the fluctuating magnetization, the full expression of the free energy has to be derived. By assuming $\gamma = \frac{\xi_{1z}^2}{\xi_{1x}^2}$ as a measure of the anisotropy between the plane and *z* axis and by neglecting the in-plane anisotropy, one writes

$$
\xi_{1,a}^2 q_a^2 = \frac{H^2}{H_{c1x}^2} \left(1 + \frac{\gamma}{2} \sin^2 \theta \right), \qquad \xi_{2x}^2 q_{\parallel}^2 = \xi_{2x}^2 (q_x^2 + q_y^2) = \frac{H^2}{H_{c2x}^2}, \qquad \xi_{2z}^2 q_z^2 = \frac{H^2}{H_{c2z}^2} \sin^2 \theta, \tag{4}
$$

where θ is the angle between *H* and the *z* axis. In MgB₂ it can be assumed [6] that $\xi_{1x} \approx \xi_{2x}$, so that the critical fields become $H_{c1x}^2 = H_{c2x}^2 = 8\phi_0^2 / \pi^2 \xi_{1x}^2 d^2$ and $H_{c2z}^2 = 16\phi_0^2 / \pi^2 \xi_{2z}^2 d^2$.

From Eqs. (1) and (2), by taking into account Eq. (4), a lengthy expression for the angular-dependent diamagnetic magnetization is obtained. For external field along the *z* axis, no double structure in the magnetization curves is present, as could be expected since only one effective correlation length is involved:

$$
M_{\text{dia}}(T = \text{const}, \theta = 0, H) = -2k_B T V H \frac{\frac{1}{H_{\text{cx}}^2} (1 + \frac{\pi^2}{2}) \epsilon + \frac{1}{H_{\text{cx}}^2} (W_{22} + \frac{\pi^2}{2} W_{11}) + \pi^2 \frac{H^2}{H_{\text{cx}}^2}}{[W_{11} + W_{22} + \frac{H^2}{H_{\text{cx}}^2} (1 + \frac{\pi^2}{2})] \epsilon + \frac{H^2}{H_{\text{cx}}^2} (W_{22} + \frac{\pi^2}{2} W_{11}) + \frac{H^4}{H_{\text{cx}}^4}}.
$$
(5)

On the contrary, for field in the *ab* plane the magnetization turns out

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$$
M_{\text{dia}}\left(T = \text{const}, \theta = \frac{\pi}{2}, H\right) = -\frac{\pi k_B T d^3 H}{3S(H)} \Biggl\{ \Biggl[\Biggl(1 + \frac{1}{2}\gamma \Biggr) + f \Biggl(\frac{\pi^2}{2}t \Biggr) + H^2 f' + H_{cx}^2 g' \Biggr] \frac{\epsilon}{H_{cx}^2} + \frac{1}{H_{cx}^2} \Biggl[W_{11} + \frac{H^2}{H_{cx}^2} (2 + \gamma) \Biggr] f \Biggl(\frac{\pi^2}{2}t \Biggr) + \frac{H^2}{H_{cx}^2} \Biggl[W_{11} + \frac{H^2}{H_{cx}^2} \Biggl(1 + \frac{1}{2}\gamma \Biggr) \Biggr] f' + \frac{1}{H_{cx}^2} \Biggl(1 + \frac{1}{2}\gamma \Biggr) g(t) + \Biggl[W_{11} + \frac{H^2}{H_{cx}^2} \Biggl(1 + \frac{1}{2}\gamma \Biggr) \Biggr] g' + \frac{1}{H_{cx}^2} \Biggl(1 + \frac{1}{2}\gamma \Biggr) W_{22} \Biggr\},
$$
(6)

where $t = \frac{2}{\pi^2} \frac{H^2}{H_{c2z}^2}$ and *g*^{*l*} and *f'* are the derivative of *g* and *f* functions with respect to H^2 . In Eq. (6) the function *S*(*H*) takes the form

$$
S(H) = W_{11} + W_{22} + \frac{H^2}{H_{cx}^2} \left(1 + \frac{1}{2} \gamma \right) + f \left(\frac{\pi^2}{2} t \right) \frac{H^2}{H_{cx}^2} + g(t) \epsilon + \frac{H^2}{H_{cx}^2} \left[W_{11} + \frac{H^2}{H_{cx}^2} \left(1 + \frac{1}{2} \gamma \right) f \left(\frac{\pi^2}{2} t \right) \right]
$$

+
$$
\left[W_{11} + \frac{H^2}{H_{cx}^2} \left(1 + \frac{1}{2} \gamma \right) \right] g(t) + \frac{H^2}{H_{cx}^2} \left(1 + \frac{1}{2} \gamma \right) W_{22}.
$$

In Fig. 2 the theoretical magnetization resulting from this equation is reported for a representative value of the reduced temperature ε and compared with the one derived for a single band for the effective correlation length. It is noticeable how the shape of the magnetization curve is affected by the two bands, displaying a double structure which resembles two upturn fields.

Single crystals of $MgB₂$ large enough to yield a size of diamagnetic signal above T_c are not available, and therefore our measurements have been carried out in powders. Thus the data reported in Fig. 1 can be considered approximately correspondent to the theoretical case of $\theta \approx \frac{\pi}{2}$. From the inset of Fig. 1 it is noted that for T_c close to T_c one has a magnetization curve similar to the dashed curve in Fig. 2. This type of field dependence, with a single upturn field, can be considered a signature of the validity of the GL regime, for $\varepsilon \approx 3 \times 10^{-3}$. Equation (3) gives a satisfactory fitting of the data in correspondence to a value

FIG. 2. Comparison of the field dependence of the fluctuating magnetization above T_c , for reduced temperature $\epsilon = 8 \times 10^{-3}$, in the case of a single superconducting band (dashed line) and for the σ and π bands (solid line).

 $\tilde{\xi} = 22$ nm, compatible with the coherence length (see Ref. [7]).

For *T* not too close to T_c , M_{dia} should be discussed in terms of the angular-dependent full expression (not reported here) by averaging over θ . By using for a qualitative illustration the form of the magnetization pertaining to $\theta =$ $\frac{\pi}{2}$, a reasonable fit of the data is obtained (Fig. 3), in correspondence with $\gamma \approx 0.02$. The critical fields turn out to be $H_{c1x} = 9.8 \times 10^4$ Oe and $H_{c2z} = 11.5 \times 10^2$ Oe, dependent on temperature through $d = \tilde{\xi}(T) = \tilde{\xi}(0) \varepsilon^{-1/2}$. It should be remarked that from the analysis of the data it turns out that the ratio W_{11}/W_{22} is temperature dependent.

FIG. 3 (color online). Fits of the magnetization curves at two representative temperatures above T_c in powered MgB₂ (see Fig. 1) on the basis of the theoretical expression [Eq. (6)] in the text, according to the generalized, nonlocal GL functional. In the inset the temperature dependence of the ratios $\frac{W_{11}}{W_{22}}$ and $M_{\text{dia}}(H_{\text{up}}^1, T)$ $\frac{M_{\text{dia}}(H_{\text{up}}^2, T)}{M_{\text{dia}}(H_{\text{up}}^2, T)}$ is reported. The departure between the theoretical prediction and the experimental data for $H \rightarrow 0$ is likely to be due to the subtraction procedure. In fact, a very small amount of ferromagnetic impurities could enhance the paramagnetic term for small fields.

In fact, W_{11}/W_{22} follows the temperature behavior of the ratio $M_{\text{up1}}/M_{\text{up2}}$ at the minimum of the magnetization curves (see the inset of Fig. 3). In the classical model [8] for the pairing mechanism leading to superconductivity, the electron-phonon interaction is generally assumed as temperature independent. This conventional assumption is made in the limit of zero temperature. In contrast, Marsiglio [9] has discussed how that assumption might be incorrect when the quasiparticle concept is extended to finite temperature. However, it seems difficult to evidence the finite temperature effect on the electron-phonon coupling constant (involved in our *W*'s) in conventional superconductors.

A simple observation on the ground of physical aspects is the following. In the presence of a finite magnetic field *H* the effective transition temperature $T_c(H)$ [which in the zero-dimensional Ginzburg-Landau approximation is $T_c(H) = T_c(0)[1 - H^2/H_c^2]$, with $H_c \tilde{\sigma} (0)^{-2}$ is different for the two bands, because of the different ξ_{σ} and ξ_{π} . When a measure is carried out at a given temperature one is in the condition of two different ε 's. By increasing temperature one has different contributions to the fluctuating magnetization at the upturn fields. This different temperature behavior is reflected in the temperature dependence of W_{11}/W_{22} .

In summary, in this work we have shown that the presence of the two superconducting σ and π bands in MgB₂ has a noticeable effect also in the fluctuating diamagnetism above T_c . By adapting to the fluctuating diamagnetism, the approach recently developed for the paraconductivity, a model extending the Ginzburg-Landau formalism to include the effect of nonevanescent magnetic field has been developed. In particular, by relying on the zerodimensional approximation for the superconducting droplets, the model has been proven to account for the basic aspects of the experimental findings.

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