Superconducting Diamagnetic Fluctuations in MgB$_2$

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The fluctuating diamagnetic magnetization $M_f$ at constant field $H$ as a function of temperature and the isothermal magnetization $M_I$ vs $H$ are measured in MgB$_2$, above the superconducting transition temperature. The expressions for $M_f$ in randomly oriented powders are derived in the Gaussian approximation of local Ginzburg-Landau theory and used for the analysis of the data. The scaled magnetization $-M_f/H^{1/2}T$ is found to be field dependent. In the limit of evanescent field the behaviour for Gaussian fluctuations is obeyed while for $H$=100 Oe the field tends to suppress the fluctuating pairs, with a field dependence of $M_f$ close to the one expected when short wavelength fluctuations and non-local electrodynamic effects are taken into account. Our data, besides providing the isothermal magnetization curves for $T > T_c(0)$ in a BCS-type superconductor such as MgB$_2$, evidence an enhancement of the fluctuating diamagnetism which is related to the occurrence in this new superconductor of an anisotropic spectrum of the superconducting fluctuations.

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Thermodynamical fluctuations on approaching the superconducting (SC) transition temperature from above yield the formation of evanescent SC droplets causing a bulk diamagnetic magnetization $-M_f$. This fluctuating diamagnetism is strongly enhanced in high temperature superconductors with respect to conventional SC’s because of the high temperature range and of the anisotropy of the cuprates. In spite of the difficulty of evidencing the superconducting fluctuations (SF) in low-temperature conventional BCS superconductors, the fluctuating diamagnetism (FD) can be detected also in these systems by means of SQUID magnetization measurements. Early data for $M_f$ at constant field as a function of temperature in zero dimensional limit (aluminum particles of size less than 1000 Å) and in metals compounds, evidenced the rounding of the transition due to SF and the effect of the magnetic field in quenching the fluctuating Cooper pairs. From the measurements of $M_f$ vs $T$ of Gollub et al. one can deduce the occurrence of an upturn in the field dependence of $M_f$: for $H \ll H_{up}$ the diamagnetic magnetization increases with $H$ while for $H \gtrsim H_{up}$ the field tends to suppress the fluctuating magnetization. The upturn field $H_{up}$ can be approximately related to the Ginzburg-Landau (GL) coherence length $\xi(T)$ (see later on). Most likely in view of the small value of $M_f$ in conventional BCS superconductors, isothermal magnetization curves $M_f$ vs $H$ have not been studied in detail, in the authors’s knowledge. The relevance of the field dependence of $M_f(T, H)$ for the study of FD has been recently stressed in the framework of a Gaussian GL approach for non-isotropic systems. The new superconductor MgB$_2$, although being of “conventional” BCS character has two characteristics that can be expected to enhance SF and therefore the value of $M_f$ at $T = T_c(0)$: the high value of the transition temperature and, as we will deduce later on, an anisotropic spectrum of the fluctuations, similarly to cuprate superconductors.

This report deals with a study of FD in MgB$_2$ by means of high resolution SQUID magnetization measurements, with a successful detection of the magnetization curves $-M_f$ vs $H$ in the temperature range $T_c(0) \lesssim T \lesssim T_c(0) + 0.5$ K. From the experimental findings we have been able to prove that $M_f$ and its field dependence are close to the one predicted by theories based on the extension of the GL approach to include short wavelength fluctuations of the order parameter and non local electrodynamic effects. It is also deduced that MgB$_2$ has an anisotropic spectrum of fluctuations, with remarkable enhancement of the FD.

The measurements have been carried out by means of the Quantum Design MPMS-XL7 SQUID magnetometer, allowing one to achieve temperature resolution up to 1 mK and the measure of a lowest magnetic moment value around $10^{-7}$ emu, in practice corresponding with our samples to the detectability of $M_f$ in field less than one Oersted. The magnetic field could be increased up to $7 \times 10^4$ Oe. The sample was prepared by Palenzona et al. (University of Genova) starting from high purity B and Mg powders, heated at 950 °C for 24 hs. A temperature-independent paramagnetic magnetization was detected in the range 100-40 K, yielding a volume Pauli susceptibility around $2 \times 10^{-7}$. The diamagnetic magnetization around $T_c$ was obtained from the raw data by subtracting the paramagnetic magnetization, as well as the temperature-independent correction due to the sample holder. Zero field cooled (ZFC) magnetization curves have been compared to field cooled (FC) data, obtained by cooling to a given temperature in the presence of different fields.
In Fig. 1 the temperature dependence of the scaled volume magnetization $m = -M_B/H^{1/2}T_c$ is reported, for MgB$_2$, and for comparison (see discussion later on), for optimally doped YBCO (oriented powder). One can deduce the following. In the temperature range indicated by a), $m$ linearly decreases on increasing temperature, corresponding to a diamagnetic susceptibility going as $(1 - T/T_c(H))$ as expected outside from the critical region below $T_c$. From this behaviour one can deduce $T_{c2}(H) = T_c(H)$ at various fields (the superconducting transition remains second order even in the presence of the field) and we derived $(dH_{c2}/dT)$ for $H \to 0$ around 800 Oe/K. In the vicinity of $T_c(H)$ the rounding of the transition due to critical fluctuations [3] originates an almost exponential temperature dependence for $m$. Because of the broadened transition region, and possibly of the powder character of the sample, the zero field transition temperature can be estimated only with a rather large uncertainty: $T_c(0) = 39.07 \pm 0.04$ K.

Now we are going to present and analyze the isothermal magnetization curves (Fig. 2). The departure from the Prange law $M_B(T = T_c) \propto H^{1/2}$ and $M_B(T \gg T_c) \propto H$, valid in the limit $H \to 0$, is evident. The upturn field $H_{up}(T)$, as well as the maximum negative value reached by $M_B$ at $H_{up}$, are functions of temperature. Let us first give a simple qualitative justification of these experimental findings, based on the assumption that the fluctuation-induced evanescent SC droplets are spherical, with a diameter of the order of the coherence length $\xi(T)$. It is noted that those droplets are the ones actually yielding the most effective diamagnetic screening [3].

For the droplets of radius $d \ll \xi(T)$ the zero dimensional approximation can be used. In this case the order parameter is no longer spatial dependent and for the magnetization an exact solution in the framework of the GL functional, valid for all fields $H \ll H_{c2}(0)$ can be found [7].

$$M_B^{D=0} = -k_BT \frac{2\pi^2 \xi(0)^2 d^2/5\Phi_0^2}{(\varepsilon + \pi^2 \xi(0)^2 H^2 d^2/5\Phi_0^2)} \cdot H$$

(1)

Now extending this result for $d \sim \xi(T)$ the upturn field turns out $H_{up} = \varepsilon \Phi_0/\xi(0)^2$, with $\varepsilon = [(T - T_c(0))/T_c(0)]$ while the magnetization at $H_{up}$ decreases on increasing temperature approximately as $M_B(H_{up}) \propto \varepsilon^{-1}$. The data in Fig. 2 are qualitatively accounted for by the above considerations.

In Fig. 3 the value of the scaled magnetization $m = -M_B(T)/H^{1/2}T$ is reported as a function of the magnetic field. It is noted that $m_c = m(T = T_c)$ decays with the field and reaches half of the value $m_c(H \to 0)$ at about $H_c = 100$ Oe. For temperature far from $T_c(0)$ and small field, the magnetization tends to the linear increase with $H$ and then $m \propto H^{1/2}$. For larger $H$ the departure from the behaviour expected in the framework of Gaussian GL theories in finite fields is dramatic. It can be observed that the field dependence of the FD in MgB$_2$ is similar to the one expected according to theories taking into account short wave length fluctuations and non local electrodynamical effects, as detected in conventional BCS superconductors [3].

Thus we first discuss the experimental findings on the basis of the fluctuation part of the Gibbs free energy of an anisotropic superconductor [4].

$$G(\varepsilon, h) = \frac{V k_BT h^{3/2}}{2^{5/2}\pi^{3/2}(0)} \zeta \left( -\frac{1}{2}, -\frac{1}{2} + \frac{h}{2H} \right), \quad h = \frac{H}{H_{c2}(0)},$$

(2)

where $\xi(0) = [\xi_{ab}(0)\xi_c(0)]^{1/3}$ here is the geometrical average of the three components of the coherence length and $H_{c2}(0)$ is angular dependent. For a randomly oriented powder we have to average with respect to the angle $\theta$ between the magnetic field and the $c$-axis of the microcrystals, resulting

$$G(\varepsilon, H) = 2\pi^{1/2}k_BT V \int_0^1 \left( \frac{g(z)H}{\Phi_0} \right)^{3/2} \zeta \left( -\frac{1}{2}, -\frac{1}{2} + \frac{\Phi_0 \varepsilon}{4\pi \xi^2(0)g(z)H} \right) dz,$$

(3)

where the anisotropy function

$$g(z; \gamma) \equiv \gamma^{-1/3} \sqrt{1 + (\gamma^2 - 1)z^2}, \quad z \equiv \cos \theta = H_z/H,$$

(4)

describes the angular dependence of the upper critical field $H_{c2} \propto 1/g(\theta)$ and $\gamma = \xi_{ab}(0)/\xi_c(0)$ is the anisotropy parameter.

The averaged magnetization can be derived by differentiation of $G$ with respect to the field [5], yielding

$$M_B(\varepsilon, h) = -\frac{1}{V} \left( \frac{\partial G}{\partial H} \right)_T = \frac{3\sqrt{\pi}}{\Phi_0^{3/2}} k_BT \sqrt{H} \int_0^1 g^{3/2}(z) \zeta \left( -\frac{1}{2}, -\frac{1}{2} + \frac{\Phi_0 \varepsilon}{4\pi \xi^2(0)g(z)H} \right) dz$$

$$+ \frac{k_BT \varepsilon}{4\xi^2(0) (\pi \Phi_0 H)^{1/2}} \int_0^1 g^{1/2}(z) \zeta \left( -\frac{1}{2}, -\frac{1}{2} + \frac{\Phi_0 \varepsilon}{4\pi \xi^2(0)g(z)H} \right) dz.$$
For the magnetization at $T_c$ one has
\[ M_\text{fl}(T_c, H) = C_0 \frac{k_B T_c}{\Phi_0^{3/2}} J(\gamma) \sqrt{H} \] with \[ C_0 = 3\sqrt{\pi} \left[ -1 + \frac{1}{\sqrt{2}} \right] \zeta \left( -\frac{1}{2} \right) = 0.32377 \] (6)

and \[ J(\gamma) \equiv \int_0^1 g^{3/2}(z; \gamma) dz = \gamma \int_0^1 \left[ \left( 1 - \frac{1}{\gamma^2} \right) z^2 + \frac{1}{\gamma^2} \right]^{3/4} dz \approx \frac{2}{5} \gamma, \quad \text{for } \gamma \gg 1, \] (7)

where $\zeta \left( -\frac{1}{2} \right) = -0.207886 \ldots$ is the Riemann zeta-function. The domain of applicability of the Prange’s law $M_\text{fl} \propto \sqrt{HT} \approx \text{const}$ can be significantly limited by critical fluctuations, anisotropy and nonlocality effects. It can be written as
\[ \frac{\pi^3 \gamma^{1/3} \mu_0^2 (k_B T_c)^2 \lambda_4(0)}{2 \Phi_0^{3/2} \xi(0)} \ll H \ll \frac{\Phi_0}{2 \pi \xi(0)^2} \gamma^{2/3}, \quad \mu_0 = 4\pi, \] (8)

while the fluctuating magnetization above $T_c$ is
\[ M_\text{fl}(T > T_c, H \to 0) = -\frac{\pi}{6} \frac{k_B T_c}{\Phi_0} \frac{\xi(0)}{\sqrt{\varepsilon}} K(\gamma) H, \quad K(\gamma) \equiv \int_0^1 g(z; \gamma) dz = \frac{2}{3} \gamma^{-2/3} + \frac{1}{3} \gamma^{4/3}. \] (9)

Let us comment on the absolute value of the fluctuating magnetization around the transition. According to the above equations and to scaling arguments [4, 10], one has
\[ m_c = \frac{M_\text{fl}(T_c)}{\sqrt{H} \cdot T_c} = \frac{k_B}{\Phi_0^{3/2}} \cdot \frac{m_D(\infty)}{k_B} = \frac{k_B}{\Phi_0^{3/2}} \cdot (-0.324) \cdot \gamma \] (10)

where for anisotropic superconductors the field should be along the c-axis (cf. Ref. 4, 5).

From the data for $m$ reported in Fig. 1 one sees that the results obtained in MgB$_2$ are consistent with a spectrum of SF with a strong enhancement factor. For a quantitative estimate, also in view of the uncertainty in the transition temperature, we preferred to compare the value of $m_c$ with the one in grain-oriented YBCO placed in magnetic field along the c-axis. It turns out that the absolute value of the scaled magnetization is slightly field dependent also in YBCO, where the crossing of $m(T)$ for different fields at $T_c(0)$ seems to be well verified [8] for $H \gtrsim 1000$ Oe. While our data coincide with the one in Ref. [4] for $H=1000$ Oe, a significative increase of $m$ appear when the field is reduced to 3 Oe. By using our data in small field and comparing the results for $m$ at $T_c(0)$ in powdered MgB$_2$ and in YBCO, where $\gamma \simeq 7$, in the light of Eq. (6) and (7) one deduces an anisotropic factor of the same order in both cases. It should be remarked that the anisotropy degree of MgB$_2$ is still uncertain, with reports giving values unexpectedly large particularly for powders, ranging from 5 to 9 [11].

In summary, by means of SQUID magnetization measurements we have experimentally detected the fluctuating diamagnetism and the magnetization curves above $T_c$ in MgB$_2$. The data have been first analyzed in terms of the theory for powdered anisotropic superconductor within the Gaussian approximation of the GL scenario. It has been found that only in the limit of evanescent field the theory is obeyed, while for $H \gtrsim 100$ Oe the field tends to suppress the fluctuating pairs and the behavior of $M_\text{fl}$ is similar to the one observed in conventional BCS superconductor and attributed to short wavelengths fluctuations and non local effects. The absolute value of the magnetization at $T_c$ indicates that the spectrum of fluctuations in MgB$_2$ is clearly anisotropic, to an extent close to the one in optimally doped YBCO.

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**FIG. 1.** Temperature dependence of the scaled magnetization $m = -M_{fl}/H^{1/2}T_c$ in MgB$_2$ around the superconducting transition temperature in a field of 1 Oe (empty squares). For comparison (see text) the data in oriented powder of optimally doped YBCO for field along the c-axis are reported, for $H = 3$ Oe (full circles) and for $H = 1000$ Oe (empty triangles), with the dotted line for the eye. The value of the scaled magnetization expected at $T_c(0)$ for isotropic 3D systems and Gaussian fluctuations, $4.7 \times 10^{-7}$, is indicated.

**FIG. 2.** ZFC isothermal magnetization curves in MgB$_2$ at typical temperatures. No difference was observed for the magnetization data in FC condition (data not reported). While in the limit $H \to 0$ one has $-M_{fl}(T \simeq T_c) \propto H^{3/2}$ and $-M_{fl}(T \gg T_c) \propto H$, for field $H \gtrsim 100$ Oe the departure from the behavior expected in the framework of Gaussian GL theories is dramatic. For a qualitative justification of the upturn field $H_{up}$ and of $-M_{fl}(H_{up})$ see text.

**FIG. 3.** Field dependence of the scaled magnetizations at typical temperatures. For $T \simeq T_c$ (squares) the Prange law $M(T_c) \propto H^{1/2}$ is verified, for $H \lesssim 50$ Gauss. For $T \gtrsim T_c + 0.2K$ (triangles and circles) one has $-M_{fl} \propto H$, again only for $H \lesssim 50$ Gauss.
\( m = - \frac{M_{fl}}{H^{1/2} T_c} \, \text{emu/cm}^3 \, \text{Oe}^{1/2} \, \text{K} \)

Fig. 1
Fig. 2

$M_{fl}$ (emu/cm$^3$) vs. $H$ (Oe)

- $H_{up} \approx 80$ Oe
- $H_{up} \approx 100$ Oe
- $H_{up} \approx 800$ Oe

Data points for:
- 39.5 K (solid circle)
- 39.3 K (up triangle)
- 39.2 K (star)
- 39.1 K (square)

Fig. 2
\[ m = -M_{fl}(T)/H^{1/2} T_c \left[ G / (Oe)^{1/2} K \right] \]

Field independent

Fig. 3