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Search for characteristic lengthscales of the magnetic field distribution at the surface of superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals

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Abstract

To probe magnetic field inhomogeneities related to vortices in superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals (T_c=90K), we have studied the EPR line of a free radical layer at heights above the surface of the order of 1000Å (the vortex lattice parameter at the free radical resonance field, 3.3kG.) For 55K<T<T_c, field inhomogeneities at the surface are on a lengthscale larger than 1000Å, a lengthscale inconsistent with a vortex lattice. Local measurements with a small DPPH grain show that a fraction of these inhomogeneities can be accounted for by the inhomogeneity of the demagnetizing field in our samples. We compare our results on the field distribution to those of μSR on similar crystals.

I. Introduction

In order to study the mixed state in superconductors, NMR[1] and μSR[2] measurements can give the overall field distribution close to the surface or in the bulk of the sample. But these techniques cannot distinguish between the intrinsic inhomogeneities, due to flux quantization and those due to the sample shape or texture. Taking into account the extrinsic sources of inhomogeneities has proven important for the determination of a reliable value for λ_L by substracting their contribution to the measured field distribution. In ceramics, with our technique, it was shown that inhomogeneities due to the grain structure account for more than half of the total field distribution[3]. Bitter-decoration experiments[4] are another way of studying the mixed state but these can only be done at low field. Our technique, based on EPR, measures field inhomogeneities at the surface of a sample around a field of 3.3kG and gives the lengthscale on which they occur. In this paper, we give the values for the first and second moments of the field distribution at the surface of $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals and make a quantitative comparison of our results with those obtained by μSR on similar samples.

II. Principle and Sample Preparation

Our method relies on the following argument. Considering a magnetic sample (z<0), the equations of magnetostatics can be solved outside the sample (z>0) given the field distribution at the surface (z=0). Assuming a distribution with a well defined lengthscale Λ along the z axis :

$$H_Z(x,y,0) = h_0 \sin px \sin qy \qquad (1)$$

where p and q are such that :

$$\Lambda = \frac{2\Pi}{\sqrt{p^2+q^2}} \qquad (2)$$

The field for z>0 is given by :

$$H_Z(x,y,z) = h_0 \sin px \sin qy \exp\left[-\frac{2\Pi z}{\Lambda}\right] \qquad (3)$$

The result is that the amplitude of the magnetic field inhomogeneities is smeared exponentially as a function of the distance from the surface, on a range which is the

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inverse of the wavevector that characterize the field fluctuations.

Our EPR based technique is the following : in absence of any EPR signal of the high-T_c phases, we place on the surface of the sample a free radical with a narrow EPR line, whose shape does not depend on temperature, and we study the shift and broadening of this line due to the magnetic field distribution. To probe field inhomogeneities with lengthscales larger than Λ, we insert a buffer layer, about $\Lambda/2\Pi$ thick, between the surface and the thinner paramagnetic layer. At our working frequency (ω_o=9.5GHz, fixed), the resonance field of the free radicals is about 3.3kG and the linewidth, about 1G. In this field, the field fluctuations related to a conventional vortex lattice are to be looked for on a lengthscale of 1000Å which is of the order of the vortex lattice parameter.

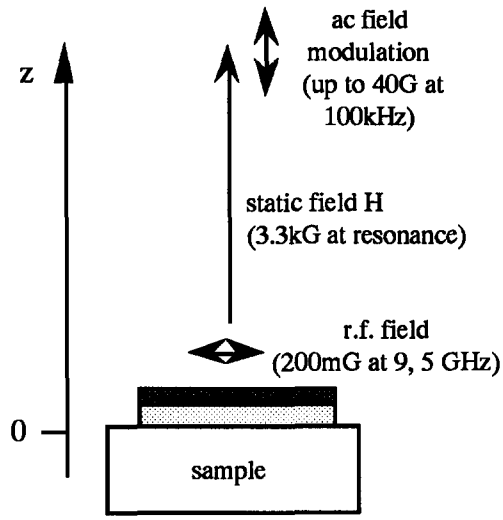

Figure 1 : geometry of the EPR measurements. The samples are prepared with a paramagnetic coating on top of a buffer layer of adjustable thickness.

We have studied single crystals of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (T_c=90K) grown by a technique described in reference [5]. These crystals have approximate dimensions of $3x3x0.05mm^3$. The surface is perpendicular to the crystalline c-axis and, observed in optical microscopy, shows no major defects except for isolated steps a few μm high. Films of hydrogen-phthalocyanin (PcH_2) of adjustable thickness (between 500 and 2000Å) for the buffer layer and lutecium-bisphthalocyanin (Pc_2Lu, 200Å thick) for the paramagnetic layer [6] were evaporated on the surface. The EPR measurements were done in the geometry shown on fig. 1 with the static field H perpendicular to the surface. The sample was field-cooled at 3.3kG and for each value of the temperature, a 100G field sweep was performed centered on the resonance field.

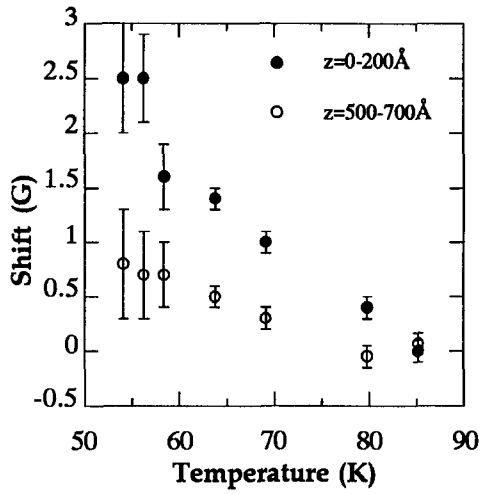

Figure 3 : Shift of the center of the resonance line (Pc_2Lu probe).

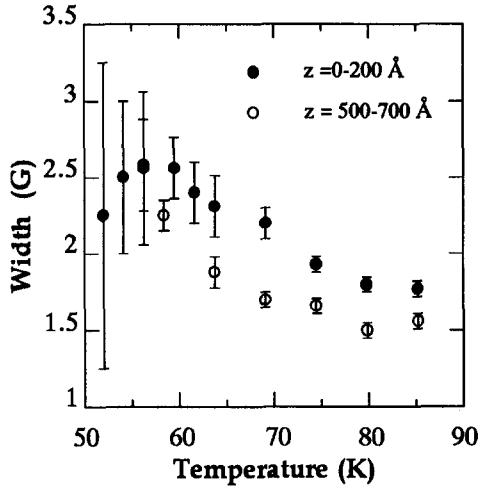

Figure 4 : Peak-to-peak linewidth of the resonance line (Pc_2Lu probe).

III. Experimental results

We present the results obtained on two crystals, one without buffer layer (z=0-200Å) and the other, with a buffer layer 500Å thick (z=500-700Å).

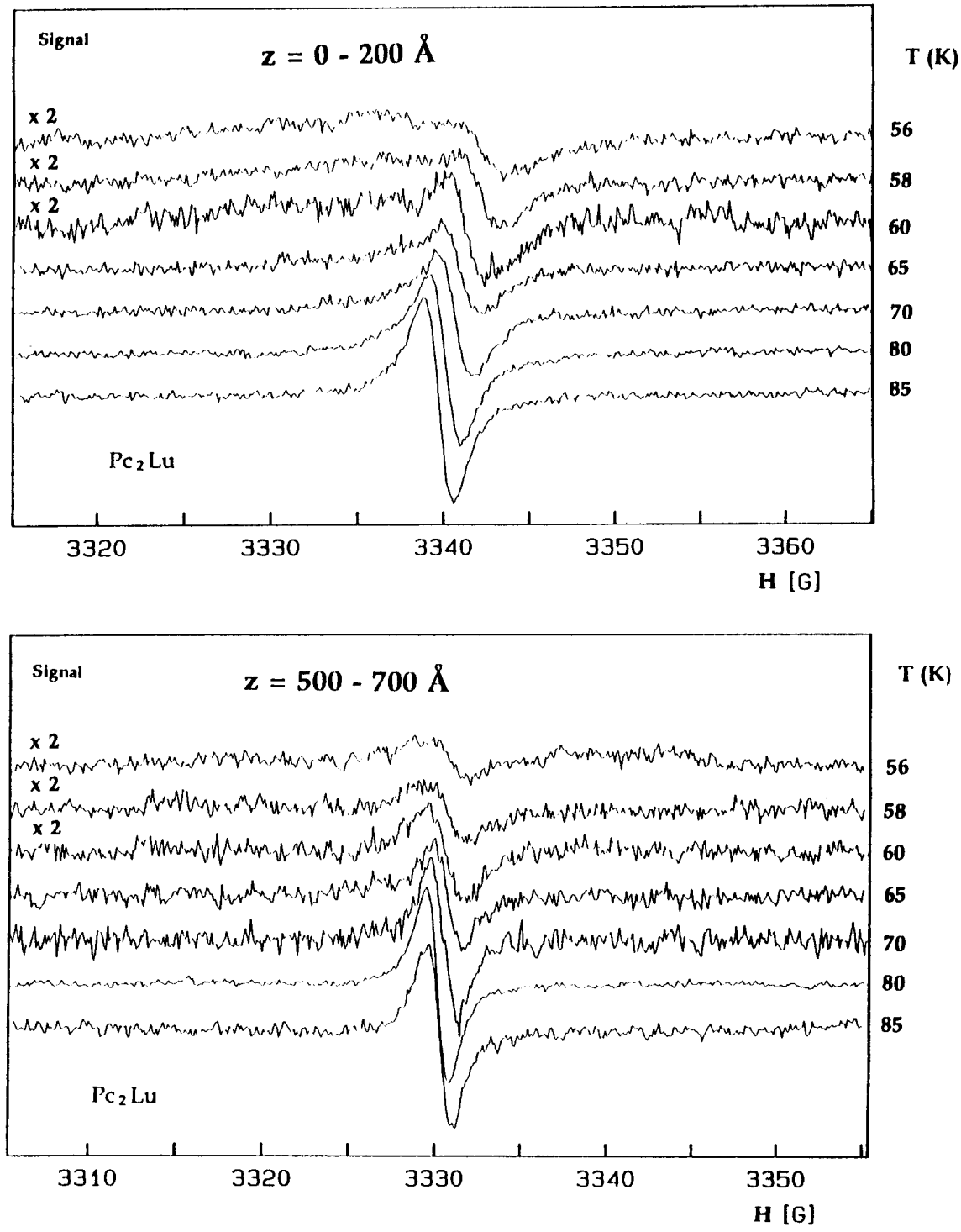

Figure 2 : EPR spectra of the 200Å thick Pc_2Lu film evaporated on the two $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals (z=0-200Å : no buffer layer; z=500-700Å : buffer layer 500Å thick).

3.1 Investigation of the microscopic field distribution

The EPR spectra, which are derivatives of the rf absorption signal due to the ac field modulation (100kHz), are presented on fig. 2 on comparable scales. In order to improve the signal to noise ratio, the spectra have been averaged over a period of 10 minutes each; the background baseline has been substracted. The Pc_2Lu layer, as the temperature decreases, behaves as an ordinary paramagnetic sample [7] down to T_c but, at lower temperature, the line shifts towards higher fields and slightly broadens; the amplitude decreases down to the limit of our sensitivity at 55K.

We have shown quantitatively that this decrease comes not only from broadening but mainly from a nonuniform macroscopic (since this behavior is common to both crystals) screening of the ac modulation [7]. Given the small volume of the probe (10^{13} spins), hence the low signal to noise ratio, we have analysed these spectra in terms of the position of the center of the line (maximum of rf absorption, fig. 3) and peak-to-peak linewidth (fig. 4).

3.2 Investigation of the macroscopic field distribution

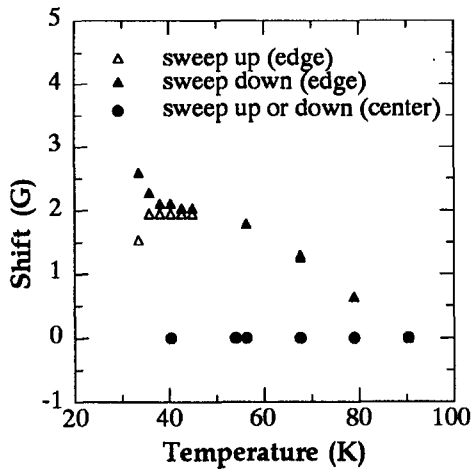

Figure 5 : Shift of the resonance line (DPPH probe).

In order to measure field inhomogeneities at the lengthscale of the dimensions of the sample, we have used a local probe however of much larger thickness than the Pc_2Lu film so that shorter lengthscales contributions are averaged out. We have placed a small DPPH grain roughly 50x50x50 μm^3 on the surface of the $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals and studied the shift of the line for different locations on the surface, typically at the center or near the edge. For each temperature, we have swept the field up and down and measured the corresponding shift for each sweep direction. The results of fig. 5 have been obtained on a typical crystal.

IV. Discussion

4.1 Modification of a resonance line due to a field distribution

In a probe containing free radicals, each spin meets the resonance condition when the value of the field is :

$$H^{res} = \frac{\omega_0}{\gamma} \qquad (4)$$

where ω_0 is the rf frequency and γ, the gyromagnetic ratio. If a local field adds up to the applied field, the value of the applied field at the resonance H^{res} will be given by:

$$H^{res} + h_{loc} = \frac{\omega_0}{\gamma} \qquad (5)$$

where h_{loc} is the local field at resonance. Let L be the intrinsic lineshape; the EPR signal S as a function of the applied field H is then :

$$S(H) = L(H - H^{res}) = L[H - (\frac{\omega_0}{\gamma} - h_{loc})] \qquad (6)$$

The total signal builds up from the sum of all signals from each spin. It can be written as :

$$S(H) = \int_{-\infty}^{+\infty} \eta(h_{loc})\, L[H - (\frac{\omega_0}{\gamma} - h_{loc})]\, dh_{loc} \qquad (7)$$

where $\eta(h_{loc})$ is the distribution of local fields at resonance in the volume of the paramagnetic probe.

In our case, the origin of $\eta(h_{loc})$ is the mixed state of the superconducting sample which affects the field outside.

4.2 Interpretation of the shift of the DPPH resonance line

Generally speaking, the shift comes from the magnetization of the superconducting sample and the flux trapped inside[8]. According to the measurements with DPPH (fig.5), the fact that there is no influence on the shift of the field sweep direction indicates that we are in a reversible regime: there is no additional pinned flux inside

the sample while the field is swept. The onset of irreversibility is clearly identified on the data of fig. 5 which displays an irreversibility temperature, T_X=(30±1)K, at 3.3kG. This determination is independent of the DPPH grain location and in good agreement with the irreversibility temperature T_{irr}(3.3kG) derived from magnetization measurements on similar crystals[9].

In the reversible regime and if we assume an average magnetization M per unit volume, weakly field dependent, the magnetic induction right below the surface is given by:

$$B^{z<0}=H+4\Pi(1-N_{loc})M \quad (8)$$

where N_{loc} is the demagnetizing factor. This factor is non uniform throughout the sample due to the non-ellipsoidal shape of the latter[10]. Given the geometry (fig. 1) and the continuity of the perpendicular component of the magnetic induction through a surface, the field where we have placed the DPPH grain is :

$$H^{z>0}=B^{z<0}= H+4\Pi(1-N_{loc})M \quad (9)$$

The position of the DPPH line is given by H^{res}:

$$H^{z>0}=\frac{\omega_0}{\gamma}=H^{res}+4\Pi(1-N_{loc})M \quad (10)$$

The shift from the position where M=0 is :

$$\Delta H_{loc}= H^{res}-\frac{\omega_0}{\gamma} = - 4\Pi(1-N_{loc})M \quad (11)$$

Since M is negative, this shift is positive. For the grain at the center (loc=c) of the crystal, we observe no shift (fig. 5), hence :

$$N_c \sim 1$$

For the grain on the edge (loc=b), we find a temperature dependant shift. As an example, at 60K:

$$\Delta H_b^{DPPH}(60K)= 1.5G = - 4\Pi(1-N_b)M(60K)$$

With N_b=0.5 [10], M(60K)= - 0.2 emu/cm^3, in good agreement with magnetization measurements on similar crystals[9].

4.3 Moments of the field distribution measured by the Pc_2Lu probe

4.3.1 First moment

According to eq. 5, the first moment $\langle h_{loc}\rangle$ is :

$$\langle h_{loc}\rangle = -\left\langle H^{res}-\frac{\omega_0}{\gamma}\right\rangle = -\langle \Delta H^{PcLu}\rangle \quad (12)$$

where $\langle \Delta H^{PcLu}\rangle$ is the average shift of the Pc_2Lu layer resonance field. This shift can be measured by the shift of the center of the line (maximum of rf absorption). This is exact only if the distribution $\eta(h_{loc})$ is symetric and provided that no spurious weighting of the field distribution comes in (such as a nonuniform ac modulation).

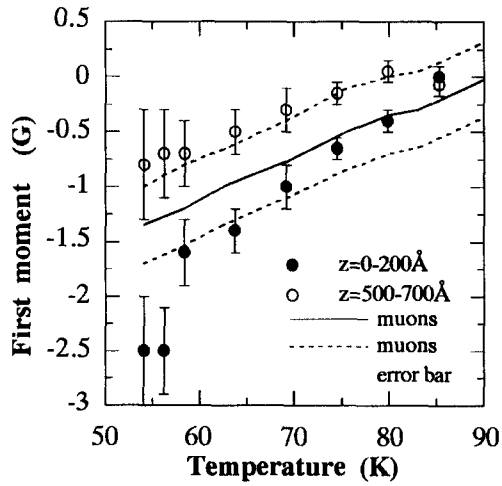

Figure 6: First moment derived from the data of fig. 3 compared to μSR results [2].

The results for z=0-200Å and z=500-700Å are presented on fig. 6. These results should be comparable to those with the DPPH probe. Combining profiles associated with the demagnetizing factor N_{loc} and with the nonuniform ac modulation [7], we have found that one should expect the following estimate for the first moment:

$$\langle \Delta H^{PcLu}\rangle \leq \frac{\Delta H_b^{DPPH}}{2}$$

Indeed, again at 60K, for z=500-700Å, the first term is 0.7G and the second, 1.5G/2. For z=0-200Å, the Pc_2Lu coating was not properly centered, thus increasing the edge effects, where the DPPH shift is maximum : this explains the larger value for the shift of this sample.

Thus, there is satisfactory agreement between all estimates of the first moment, including μSR results (fig. 6). This indicates that, in the bulk or at the surface, the origin of this negative first moment of the field distribution is the nonuniform demagnetizing field in the plane of the sample.

4.3.2 Second moment

We have derived an estimate for the second moment from the peak-to-peak linewidth. In eq. 7, we have taken

for $\eta(h_{loc})$ a gaussian distribution of second moment σ^{surf}, function of temperature. The intrinsic lineshape of Pc_2Lu, L, can be identified with a lorenztian, the peak-to-peak width being 1.7G for z=0-200Å and 1.5G for z=500-700Å, independent of temperature (the difference in width arises presumably from the fact that for z=0-200Å, Pc_2Lu was evaporated directly on the surface of the crystal). Therefore, we analyse our signals as convolutions of a lorenztian and a gaussian, a shape known as the Voigt Profile[11].

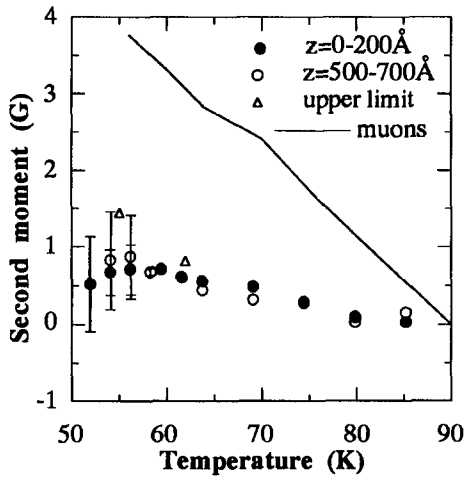

Figure 7: Second moment derived from the data of fig. 4 compared to μSR results [2].

For the two samples and for each temperature T, we evaluate the second moment $\sigma^{surf}(T)$ of the gaussian giving rise to the measured peak-to-peak width when convoluted with the intrinsic lorentzian line. This procedure may underestimate the broadening when the signal to noise ratio becomes poor, especially at T<60K. We have determined, independantly, an upper bound for the peak-to-peak width by increasing drastically the modulation [7].

The results are presented on fig. 7. The Pc_2Lu film at z=500-700Å probes the same field distribution than the one at z=0-200Å except for inhomogeneities at lengthscales below 3000Å (2Πx500Å). The results show that the second moment of the fitted gaussian is, within our experimental incertainty, the same at z=0-200Å and z=500-700Å. Thus, the field inhomogeneities at the surface are mainly on lengthscales greater than 1000Å.

A fraction of the second moment that we measure can be accounted for by the inhomogeneity of the demagnetizing field. Taking the same profiles as mentioned above for both N_{loc} and ac modulation, this fraction is :

$$\sigma^{surf}_{dem} < \frac{\Delta H_b^{DPPH}}{2\sqrt{3}}$$

At 60K, we have σ^{surf}=0.7G and σ^{surf}_{dem}<0.4G. The contribution σ^{surf}_{int} to the second moment on intermediate lengthscales, between 1000Å and 100μm, is given by :

$$(\sigma^{surf}_{int})^2 + (\sigma^{surf}_{dem})^2 = (\sigma^{surf})^2 \quad (13)$$

which gives, at 60K, σ^{surf}_{int}>0.6G.

Taking λ_{ab}=2000Å, the second moment at 60K for a conventional, static lattice of straight vortex lines should be 30G[12]. Consistently with the largest possible difference between the z=0-200Å and z=500-700Å data on fig. 7, we find less than 100mG on the lengthscale of the vortex lattice. The muons, on all lengthscales, find at most 3.5G. Therefore, both experiments converge to show that, in the temperature range 55K<T<T_c and in a field of roughly 3.3kG, the field distribution is much narrower than expected. Motional narrowing of the EPR line would require fluctuations on a timescale of 10^{-10}s (our time constant is the inverse of the unnarrowed linewidth), which seems unlikely.

The quantitative difference between our results and those of μSR can be explained, in part, by the fact that our measurement is a surface measurement and the muons, a bulk measurement. This has not the same effect on the first and second moments. For the former, the relevant geometry of the sample is the thin slab which gives a unifom demagnetizing factor along the z axis[10], hence the good agreement of our results with μSR. On the contrary, the second moment reflects primarily, as shown by the small value for σ^{surf}_{dem} at 60K, a contribution to the field distribution on lengthscales smaller than 100μm, the order of magnitude for the sample thickness. The relevant geometry in this case is a semi-infinite sample. The net effect of the surface in this geometry is to divide by a factor of 2 the local variations of the field at the surface compared to the bulk (the local field is created by currents flowing in all the space for the bulk situation and only in half of the space in the surface situation), hence :

$$\sigma^{bulk}_{int} \approx 2\,\sigma^{surf}_{int}$$

where σ^{bulk}_{int} is the second moment in the bulk of the field distribution that we probe at the surface.

This factor 2 is still not large enough to explain the difference between our results and μSR. One could think of the Pc_2Lu film of z=0-200Å being at a greater distance than 200Å from the surface. In this case, the short lengthscales contributions (1000Å) would be averaged out. This would require a 300Å thick pollution layer on the surface of the crystals prior to evaporation, which is unlikely. It could be that the muons measure field inhomogeneities at even shorter lengthscales than 1000Å. Such a possibility has been considered by Brandt in the case of dot vortices[13]. The larger lengthscales (>1000Å) that are probed by our technique can be taken as an indication of disorder of vortex dots on such lengthscales.

Conclusion

There seems to be no well defined lengthscale for the field distribution in $Bi_2Sr_2CaCu_2O_{8+\delta}$ in a field of roughly 3.3kG and in the temperature range $55K<T<T_c$. Our measurements and the above discussion suggest a highly disordered mixed state, perhaps made of randomly positioned vortex dots.

Acknowledgements

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