

Vortex lattice and vortex dynamics in $\text{YBa}_2\text{Cu}_4\text{O}_8$ from ^{89}Y NMR

Pietro Carretta*

Department of Physics "A. Volta," University of Pavia, Pavia, Italy

(Received 5 December 1991)

^{89}Y NMR spectra in oxygen-deficient, pure and calcium-doped $\text{YBa}_2\text{Cu}_4\text{O}_8$ powdered samples below T_c are presented. The square root of the derived second moment of the field distribution inside the vortex lattice at low temperatures appears to be linearly related to T_c . The values derived for $\langle \Delta B^2 \rangle$, however, are remarkably different from the ones obtained from $\mu^+\text{SR}$. At higher temperatures a narrowing of the lines is observed and a crossover between two different temperature dependences of the vortex dynamics is evidenced.

The study of the vortex lattice structure and of the vortex line dynamics is one of the most relevant issues in the field of high- T_c superconductivity. The understanding of the dependence of vortex lattice structure on extrinsic parameters, such as the number of pinning centers or the strength of the external magnetic field, is of crucial importance in view of the relevant quantities that can be derived, the penetration depth λ for instance. On the other hand, through the study of the thermally induced vortex dynamics the strength of the coupling, Josephson or simply magnetic, between adjacent CuO_2 layers can be extracted. Until now the vortex structure has been investigated mostly through $\mu^+\text{SR}$ (Ref. 1) or macroscopic techniques,² while vortex dynamics has been studied, almost exclusively, by means of techniques such as dynamical susceptibility, resistivity measurements, and magnetization relaxation experiments.³

A microscopic probe to investigate the structure of the vortex lattice is NMR. Through the detailed analysis of the line shapes, insights on the field distribution inside the vortex lattice, and estimates of the penetration depth λ can be obtained.⁴ Recently such an approach has been applied by means of ^{89}Y NMR below T_c in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$.^{5,6} Since the ^{89}Y nucleus is not sensitive to the electric-field gradient, it is, in most respects, an ideal tool to probe the magnetic-field distribution. Besides providing information on the vortex lattice structure the analysis of a ^{89}Y NMR has evidenced a narrowing related to vortex motions.⁵ Thus, a direct probe of the microscopic vortex dynamics is available.

In this paper a study of ^{89}Y NMR spectra in oxygen-deficient, calcium-doped and pure $\text{YBa}_2\text{Cu}_4\text{O}_8$ (Y 1:2:4) superconductors below T_c is presented in order to extract information on the vortex lattice through the analysis of the ^{89}Y NMR linewidth $\Delta\nu$ and through its motional narrowing. In the low-temperature range the derived second moment of the field distribution $\langle \Delta B^2 \rangle$ is compared with the one obtained from $\mu^+\text{SR}$.¹ For $T \rightarrow T_c^-$ a narrowing of the line is observed and a crossover between two different temperature dependences of the vortex dynamics is evidenced.

The transition temperature of our powder samples was derived either from Meissner effect measurements or from magnetically modulated radio-frequency absorption experiments.⁷ One has for $\text{YBa}_2\text{Cu}_4\text{O}_{7.9}$, $T_c = 68 \pm 1$ K; for

$\text{YBa}_2\text{Cu}_4\text{O}_{7.95}$, $T_c = 76 \pm 1$ K; and for $\text{YBa}_2\text{Cu}_4\text{O}_8$ 5% calcium-doped, $T_c = 84 \pm 1$ K. The irreversibility temperature, which corresponds to the temperature where a macroscopic technique starts to observe a dissipation arising from the long-wavelength motion of vortex lines, was inferred from the onset in the detuning of the NMR probe. One can estimate $T_{\text{irr}} \approx 45$ K for all the three samples. The effect of the external magnetic field ($H_0 \approx 5.9$ T) on the transition temperature of our samples was not measured. No sizable reduction of T_c is expected in view of the very steep slope of $H_{c2}(T)$ in these strongly type-II superconductors.⁸ All ^{89}Y NMR spectra were obtained from the Fourier transform of the decay of the echo after a $(\pi/2)_x - \tau - (\pi)_y$ pulse sequence, with a radio-frequency field strong enough (≈ 85 G) to irradiate the whole line. Above T_c only one ^{89}Y NMR line with $\Delta\nu \approx 1.1$ KHz was observed, in all the samples. No signal from $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ or any other phase was detected, either from ^{89}Y NMR or from ^{63}Cu nuclear quadrupole resonance (Ref. 9), evidencing the high degree of purity of the samples.

First the results of the ^{89}Y NMR spectra at low temperatures will be presented. For $T \ll T_c$ the vortex lattice can be considered rigid, namely, the characteristic correlation times of vortex motions are longer than the inverse of the intrinsic linewidth. Since in the NMR high-field regime, for the average separation d between vortices one has $\xi(0) \ll d \ll \lambda(0)$, where ξ is the coherence length, the London approximation should be valid to describe the magnetic-field distribution inside the vortex lattice.¹⁰ Moreover, due to the high anisotropy ratio between the effective masses of the superconducting carriers [$(m_c/m_{ab})^{1/2} > 5$], a simple relation connects the second moment of the field distribution expected when $\mathbf{H}_0 \parallel \mathbf{c}$ with that from the randomly oriented grains of a powder sample.¹¹ One has

$$\langle \Delta B^2 \rangle = k \frac{\Phi_0^2}{\lambda_{ab}^4} \propto \left(\frac{n}{m_{ab}} \right)^2, \quad (1)$$

where n is the concentration of superconducting carriers, λ_{ab} the penetration depth for $\mathbf{H}_0 \parallel \mathbf{c}$, and k a constant given by $k \approx 0.00162$.

The field distribution from powder samples should be anisotropic.¹¹ However, the distribution of demagnetiza-

tion factors from the variously shaped grains induces a further smearing out and the observed ^{89}Y NMR line shapes are practically close to Gaussian ones (see Fig. 1). In order to extract relevant information on the field distribution for $T \rightarrow 0$ the lines were fitted with a Gaussian function and the full width at half intensity $\Delta\nu$ was derived. $\Delta\nu$ is connected to the second moment of the field distribution by the equation $\Delta\nu = 2.36\langle\Delta B^2\rangle^{1/2}\gamma_{89}$. In Fig. 2 the transition temperature T_c is reported as a function of $\langle\Delta B^2\rangle^{1/2}$. The values for $\langle\Delta B^2\rangle^{1/2}$ appear linearly related to T_c . This proportionality confirms the findings from $\mu^+\text{SR}$ experiments¹² and implies that $T_c \propto n/m_{ab}$ [see Eq. (1)]. In the Ca-doped sample calcium seems to preferentially substitute Ba ions;¹³ therefore the substitution should not introduce any change in the number of carriers.¹⁴ Since a Ca ion is rather smaller than a Ba ion, the substitution can increase the pressure on CuO_2 layers, giving rise to a lowering of the effective mass m_{ab} and to an increase in $\langle\Delta B^2\rangle$. In the case of the oxygen-deficient sample no variation in the effective mass is expected, and the decrease of $\langle\Delta B^2\rangle$ should be related to a decrease of n , as in oxygen-deficient $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ superconductors.¹²

The second moment of the field distribution derived from $\Delta\nu$ for $T \rightarrow 0$ can be compared with the one obtained from $\mu^+\text{SR}$ experiments through the muon depolarization rate σ ($\sigma = 2\pi\gamma_\mu\langle\Delta B^2\rangle^{1/2}/\sqrt{2}$, with $\gamma_\mu = 135.5 \text{ MHz/T}$).¹² A remarkable difference between the values of $\langle\Delta B^2\rangle$ estimated from the two techniques is observed. The values from ^{89}Y NMR are much smaller than those from $\mu^+\text{SR}$

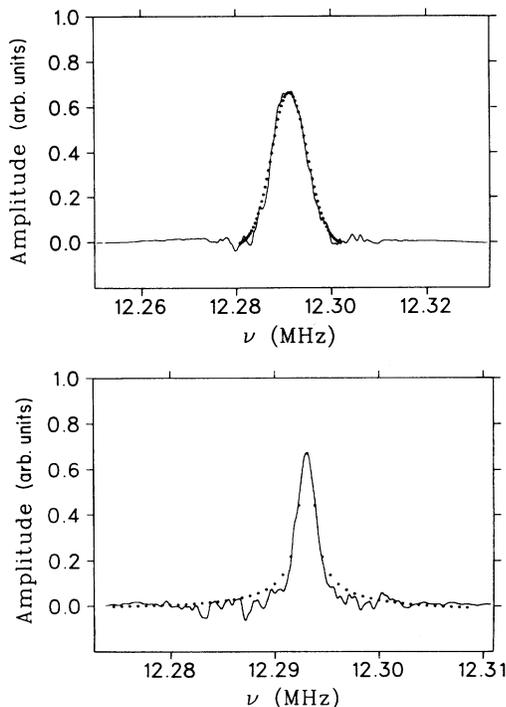


FIG. 1. ^{89}Y NMR spectra in $\text{YBa}_2\text{Cu}_4\text{O}_{7.95}$ ($T_c = 76 \text{ K}$) at $T = 18 \text{ K}$ (top) in comparison with the best-fit Gaussian function, and at $T = 68 \text{ K}$ (bottom) with the best-fit function obtained from the Fourier transform of Eq. (2) in the text, with $\tau_c = 35 \mu\text{s}$.

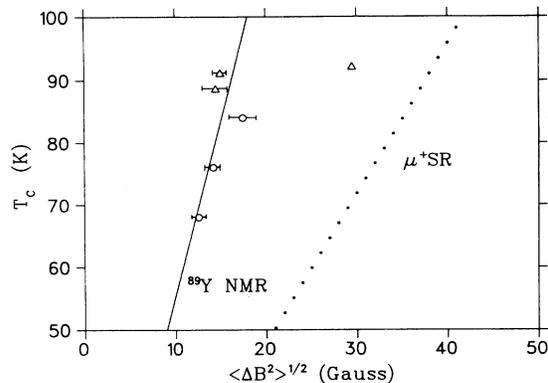


FIG. 2. Dependence of T_c from $\langle\Delta B^2\rangle^{1/2}$ for $T \rightarrow 0$, as obtained from the ^{89}Y NMR linewidth. The circles give the data extracted from our measurements in Y 1:2:4 powder samples, while triangles refer to the values derived from ^{89}Y NMR in oriented samples (Ref. 11) of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Refs. 5, 6, and 20). The solid line is the best-fit line according to the linear relation $T_c = \alpha\langle\Delta B^2\rangle^{1/2}$ with $\alpha = 5.55 \text{ K/G}$. From $\mu^+\text{SR}$ one would derive $\alpha_\mu = 2.4 \text{ K/G}$ (Ref. 12) (dotted line).

(see Fig. 2) and the slope of the best-fit linear relation line T_c vs $\langle\Delta B^2\rangle^{1/2}$ is larger by a factor of ≈ 2.3 for ^{89}Y NMR data. Although $\mu^+\text{SR}$ and NMR experiments for H_0 in the ab plane appear to probe the same field distribution,¹⁵ one should conclude that the two techniques probe different field distributions particularly when $\mathbf{H}_0 \parallel c$. The origin of this effect can be related to the large difference between the strength of the magnetic fields used in the two experiments. In $\mu^+\text{SR}$, in fact, H_0 is smaller than in NMR by a factor of about 100. When a small number of vortices is present, the vortex lines are almost all trapped by pinning centers, since the energy gain per unit length obtained by trapping the flux lines, $U_{\text{pin}} \sim (H_c^2/8\pi)a_0^2$, where $a_0 \sim d$ is the characteristic diameter of the vortex, is much larger than the repulsive interaction energy between vortices $U_{\text{rr}}(r) \sim (\Phi_0^2/8\pi\lambda^2)\ln(\lambda/r)$ (r is the separation between vortices). Therefore, in the conditions in which $\mu^+\text{SR}$ experiments are done the vortex lattice is expected to be more distorted than in the high-field NMR region, where the higher density of vortices leads to a lower percentage of pinned vortex lines and to a more regular vortex lattice with a field distribution characterized by a second moment with a lower effective constant k [see Eq. (1)]. On the contrary, for H_0 in the ab plane the number of intrinsic pinning centers between CuO_2 planes is high compared to the number of vortices, regardless of the strength of the field. Therefore no relevant difference is expected between $\mu^+\text{SR}$ and NMR findings.

Now the behavior of ^{89}Y NMR lines in the high-temperature range ($T \rightarrow T_c^-$) will be considered. By increasing the temperature the characteristic correlation time of vortex lines dynamics decreases and a narrowing of $\Delta\nu$, with respect to the behavior expected on the basis of the temperature dependence of $\lambda(T)$, is observed (see Figs. 3 and 4). As appears from the figures, two qualitatively different temperature dependences of $\Delta\nu$ vs T can be distinguished in the narrowing regime.

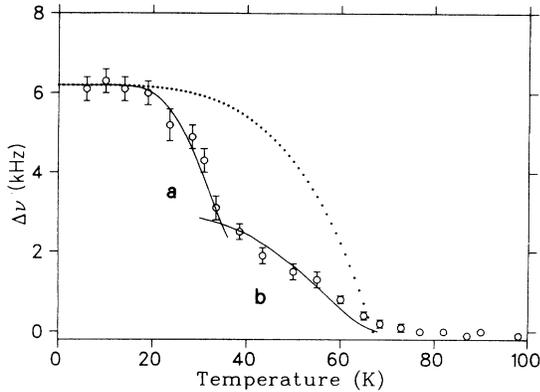


FIG. 3. Temperature dependence of the extrabroadening of the ^{89}Y NMR line below T_c for the oxygen deficient Y 1:2:4 sample (a constant value of $\Delta\nu=1.1$ KHz, the linewidth measured above T_c , has been subtracted). The dotted line shows the expected behavior for $\Delta\nu$ in the absence of vortex motion, by taking into account only the temperature variation of $\lambda(T)$. Line (a) shows the behavior of the linewidth in the low-temperature thermally activated regime for a temperature dependence of the correlation time given by $\tau_c = e^{145/T}$ μs . Line (b) shows the behavior in the high-temperature melted regime with $\tau_c = 35$ μs . $T_{\infty} \approx 35$ K is the temperature where the crossover is observed to occur.

In order to extract quantitative values for the characteristic correlation time of vortex motion τ_c , one can use the approximate expression for the free-induction decay in the case of a random Brownian motion:^{5,16}

$$s(t) = \exp\{-\gamma^2 \tau_c^2 \langle \Delta B^2 \rangle [\exp(-t/\tau_c) - 1 + t/\tau_c]\}. \quad (2)$$

By fitting the ^{89}Y NMR lines with the Fourier transform of the above expression (see Fig. 2) and by assuming a two-fluid model temperature dependence for the rigid lattice second moment $\langle \Delta B^2 \rangle$,¹ an evaluation of the temperature dependence and of the values of τ_c is derived. Thus, it is possible to compare the behaviors of τ_c in the two narrowing regimes with the ones expected from theoretical models.

In the low-temperature regime (see Figs. 3 and 4) the behavior of $\Delta\nu(T)$ supports a model of thermally activated vortex lines motion with a characteristic correlation time given by $\tau_c = \tau_0 e^{(U/k_B T)}$, where U is an effective activation energy and τ_0 is the characteristic hopping time among pinning centers. In particular, it is observed that in this regime the effective pinning potential U increases as T_c increases (see Figs. 3 and 4): $U \approx 145$ K for the $T_c = 68$ K sample, $U \approx 175$ K for the one with $T_c = 76$ K, and $U \approx 245$ K for the Ca-doped one with $T_c = 84$ K. A high effective pinning potential can be related to a strong correlation between adjacent CuO_2 planes. Therefore the above results appear to support the idea that T_c is related to the coupling between the CuO_2 layers.¹⁷

At higher temperatures, the thermal fluctuations become strong enough to overcome the weak Josephson and magnetic coupling between the CuO_2 layers and a crossover to a nonactivated (τ_c constant) regime is expected, since when the coupling between adjacent CuO_2 planes is

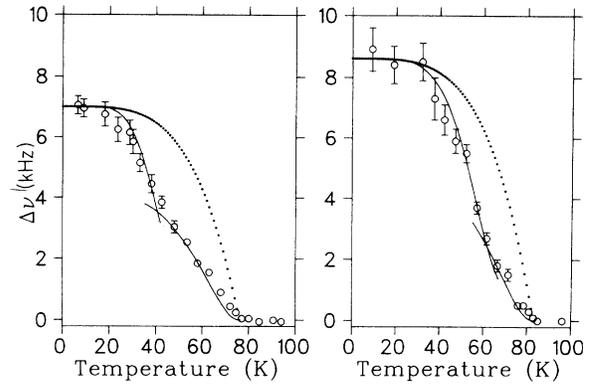


FIG. 4. Temperature dependences of the extrabroadening of the ^{89}Y NMR lines below T_c for the pure (left) and Ca doped (right) Y 1:2:4 samples. The dotted lines show the expected behaviors for the rigid lattice linewidth. The solid lines show the behaviors of the linewidths in the low-temperature thermally activated regime, where $\tau_c = e^{175/T}$ μs for the pure Y 1:2:4 sample and $\tau_c = e^{245/T}$ μs for the Ca doped one, and in the high-temperature liquidlike regime where $\tau_c = 35$ μs for both samples.

broken the single pinning center in a layer becomes less efficient because it can trap only those vortices inside the same plane and not the whole vortex line as at lower temperatures.

This idea is well supported by our data since the temperature behavior of $\Delta\nu$ in the high-temperature regime is consistent with a constant correlation time for vortex motion (see Figs. 3 and 4). The value for τ_c is the same for all samples, namely, $\tau_c \approx 35$ μs . This result is consistent with a picture of a quasi-two-dimensional liquidlike diffusion of vortices in CuO_2 planes once the coupling between these layers has been overcome,¹⁸ with an average distance between vortices which is the same for all three samples, where H_0 is the same.

In the weak Josephson-coupling limit the dominant coupling mechanism is magnetic.¹⁸ In this approximation an expression for the temperature at which the crossover from the activated to the nonactivated regime occurs, $T_{\infty} = \Phi_0^2 s / 32 \pi^2 k_B \lambda_{ab}^2$ (Ref. 18) (s is the separation between CuO_2 layers) is obtained. It must be remarked that the above expression implies that $T_{\infty} \propto \langle \Delta B^2 \rangle^{1/2}$ [see Eq. (1)], which is itself proportional to T_c , as previously observed. Although the value of the temperature at which this crossover occurs cannot be precisely determined from the ^{89}Y NMR data for $\Delta\nu(T)$, it can be noticed that T_{∞} increases with T_c (Figs. 3 and 4), in qualitative agreement with a picture where Josephson coupling between adjacent CuO_2 layers is neglected. T_{∞} does not appear related to the irreversibility temperature. On the other hand, the latter is not an intrinsic characteristic of the material, depending on the time scale of the technique used to determine it.³

In conclusion a dependence on the technique (^{89}Y NMR or $\mu^+\text{SR}$) of the extracted second moment of the magnetic-field distribution inside the vortex lattice for $T \rightarrow 0$ was observed. This phenomenon has been related to the different distortions induced by extrinsic pinning

centers on the vortex lattice when magnetic fields of very different strength are used. Moreover, a proportionality between T_c and $(\Delta B^2)^{1/2}$ was observed, in agreement with Uemura's findings from μ^+ SR.¹² At higher temperatures a narrowing of ^{89}Y NMR line is observed and it is attributed to the onset of relatively fast vortex motion. The narrowing is well described by the superposition of a low-temperature activated regime and a high-temperature nonactivated one, in agreement with the observations from magnetization relaxation experiments in $\text{YBa}_2\text{Cu}_3\text{O}_7$ superconductors.¹⁹ Estimates of the characteristic correlation time of vortex motion indicate that the effective activation energy, in the low-temperature regime, increases on increasing T_c . In the high-temperature re-

gime τ_c is not related to T_c . The derived values of τ_c are of the order of tens of microseconds, too long to allow one to detect a narrowing through μ^+ SR, where $\tau_c > (\Delta v_\mu)^{-1}$ even for T close to T_c .

Thanks are due to S. Aldrovandi and M. Corti for their technical support. C. Bucci, R. De Renzi, G. Guidi, and A. Rigamonti are gratefully thanked for useful discussions. The research has been partly supported by INFN (Istituto Nazionale di Fisica Nucleare), by INFN-GNSM (Grant No. MPI 40 %) and by the National Research Council of Italy (CNR) under the "Progetto Finalizzato Superconductive and Cryogenic Technologies."

*Also with the Department of Physics, University of Parma, Parma, Italy.

¹R. De Renzi, *Proceedings of the International Conference on Magnetism, Edinburgh, 1991* [J. Magn. Magn. Mater. (to be published)]; H. Keller, in *Earlier and Recent Aspects of Superconductivity*, edited by J. Bednorz and K. A. Müller (Springer-Verlag, Berlin, 1990), p. 222.

²K. E. Gray and D. H. Kim, *Physica C* **180**, 139 (1991).

³A. P. Malozemoff *et al.*, *Phys. Rev. B* **38**, 7203 (1988); A. P. Malozemoff, in *Physical Properties of High Temperature Superconductors*, edited by D. M. Ginsberg (World Publishing, Singapore, 1989), pp. 71–150; T. R. Chien *et al.*, *Phys. Rev. Lett.* **66**, 3075 (1991); D. Fiorani, A. M. Testa, and G. Calestani (unpublished).

⁴M. Mehring, in *Earlier and Recent Aspects of Superconductivity* (Ref. 1).

⁵P. Carretta and M. Corti, *Phys. Rev. Lett.* **68**, 1236 (1992).

⁶H. B. Brom and H. Alloul, *Physica C* **177**, 297 (1991).

⁷P. Orlandi and A. Rigamonti, *Physica C* **178**, 197 (1991).

⁸U. Welp *et al.*, *Phys. Rev. Lett.* **62**, 1908 (1989).

⁹P. Carretta *et al.*, *Physica C* **191**, 97 (1992).

¹⁰E. H. Brandt, *Phys. Rev. B* **37**, 2349 (1988); E. H. Brandt and

A. Seeger, *Adv. Phys.* **35**, 189 (1986).

¹¹W. Badford and J. M. F. Gunn, *Physica C* **156**, 515 (1988).

¹²Y. J. Uemura *et al.*, *Phys. Rev. Lett.* **62**, 2317 (1989).

¹³E. Kaldis *et al.*, in *Structural and Physical Properties of the Single and Doubled Chain Y-Ba-Cu-O Phases 124 and 123.5*, Proceedings of the M2S-High-Temperature Superconductors III Conference, Kanazawa, Japan, 1991 [*Physica C* (to be published)].

¹⁴M. Knapfer *et al.*, *Physica C* **182**, 62 (1991).

¹⁵Data reported in Ref. 5 has been compared with those from μ^+ SR reported for the same sample by L. Albanese *et al.*, in *Proceedings of the International Conference on Magnetism* (Ref. 1).

¹⁶A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961), p. 439.

¹⁷M. Di Stasio, K. A. Müller, and L. Pietronero, *Phys. Rev. Lett.* **64**, 2827 (1990).

¹⁸K. H. Fischer, *Physica C* **178**, 161 (1991); J. R. Clem, *Phys. Rev. B* **43**, 7837 (1991).

¹⁹Z. J. Huang *et al.*, *Physica C* **176**, 195 (1991).

²⁰S. E. Barrett *et al.*, *Phys. Rev. B* **41**, 6283 (1990).