Low-temperature magnetism of Nd_2CuO_4: An ultrasonic investigation

P. Richard,[1,2,*] M. Poirier,[1] and S. Jandl[1]

[1]*Regroupement Québécois sur les Matériaux de Pointe, Département de Physique, Université de Sherbrooke, Sherbrooke, Canada, J1K 2R1*

[2]*Department of Physics, Boston College, Chestnut Hill, Massachusetts 02134, USA*

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We have performed an ultrasonic study of Nd_2CuO_4 in order to investigate the low temperature magnetism due to the Nd^{3+} moments. In addition to small anomalies corresponding to the Cu^{2+} spin reorientations observed at 33 and 67 K, a large magnetic field and frequency dependent anomaly is detected around 4 K in the C_{66} elastic constant and the corresponding attenuation variations. The frequency dependence of this anomaly, which disappears under a magnetic field of 2 T applied along the in-plane Cu-O bonds, is related to a resonant phenomenon rather than to a phase transition as previously reported. The ultrasonic measurements suggest the development of local domains, due to a frustration of the magnetic structure caused by the competition between the Nd^{3+}-Cu^{2+} and the Nd^{3+}-Nd^{3+} interactions at low temperature. In contrast to the common belief, the C_{66} elastic constant and attenuation anomalies above 2 T characterize a field-induced transition of the Nd^{3+} spins which occurs at critical field lower than the one associated with the Cu^{2+} spins.

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I. INTRODUCTION

In order to clarify the role played by magnetism in the high-temperature superconductors, much effort has been devoted in the past years to the understanding of the 2-1-4 electron-doped superconductors ($RE_{2-x}Ce_xCuO_4$, RE=Pr, Nd, Sm) magnetic properties. In contrast to the hole-doped materials, for which a low concentration of holes suppresses the Cu^{2+} antiferromagnetic (AF) order, the AF order persists in $Nd_{2-x}Ce_xCuO_4$ for dopings as high as the optimal cerium concentration $x=0.15$.[1] Furthermore, recent experiments have established the AF order as a competing ground state in the electron-doped superconductors.[2] Among the 2-1-4 electron-doped superconductors, Nd_2CuO_4 has aroused a lot of interest owing to the strongly coupled Cu^{2+} and Nd^{3+} magnetic subsystems, as illustrated by the spin reorientations found around 33 and 70 K, well below the Cu^{2+} Néel temperature (~250 K).[3–6] Several experimental techniques have been used for their study, such as crystal-field infrared transmission,[7] Raman scattering,[8] muon spin relaxation,[4] and neutron measurements.[5,9,10]

As the temperature is decreased, Nd_2CuO_4 exhibits at least three magnetic phases called phase I ($70\ K \lesssim T \lesssim 250\ K$), phase II ($33\ K \lesssim T \lesssim 70\ K$), and phase III ($T \lesssim 33\ K$); the magnetic structure being the same in phases I and III. In between two consecutive phases, each three-layer-spin unit, formed by one Cu^{2+} layer sandwiched between two Nd^{3+} layers, rotates as a whole by 90° in the *ab* plane, with the adjacent unit rotating in opposite direction. While the magnetic structure of Nd_2CuO_4 is noncollinear in zero applied field,[11–13] the magnetic structure becomes collinear under a magnetic field of 4.4 and 0.75 T applied along the [100] and the [110] directions, respectively.[11] As shown in a thermal conductivity study of Nd_2CuO_4, these field-induced transitions are accompanied by the closure of a ~0.3 meV gap in an acoustic magnon branch at $\mathbf{k}=0$, leading to magnon heat transport at temperatures as high as 18 K.[14]

Even though Nd^{3+} spins are involved in these magnetic transitions, a spontaneous ordering of the Nd^{3+} subsystem at low temperature, due to a Nd^{3+}-Nd^{3+} interaction, remains controversial. Hence, while x-ray magnetic scattering data indicate that Nd^{3+} ions are polarized at 37 K,[15] the removal of the Kramers doublet degeneracies, as observed by crystal-field infrared transmission, indicates that these ions are already polarized by the Cu^{2+} subsystem at a temperature as high as 140 K.[7] An enhancement of neutron scattering magnetic peak intensities around 3 K has been interpreted by Matsuda *et al.* as a direct Nd^{3+}-Nd^{3+} interaction,[5] while Lynn *et al.* has estimated the Nd^{3+} ordering temperature around 1.5 K.[6]

The ultrasonic technique is known to be very sensitive to the magnetic structure of crystals. Hence, in a previous ultrasonic study of Nd_2CuO_4 at a frequency of 54.3 MHz, anomalies found around 33.5 K on the C_{66} and the C_{11}-C_{12} elastic moduli have been associated with the phase II→phase III magnetic transition.[16] Moreover, this study has revealed a strong anomaly (~5%) around 5 K in the C_{66} elastic constant, accompanied by a peak in the attenuation. While no frequency dependence of this anomaly has been reported, a shift towards the low temperatures, as well as a decrease in its amplitude up to the maximum magnetic field used (3 T), have been observed under a magnetic field applied along the [100] direction.[16] Weaker anomalies (<0.1%) have also been measured at the same temperature in the C_{11} and C_{44} elastic moduli. These anomalies around 5 K were attributed to a spontaneous ferroelastic phase transition, whose origin was later associated with a loss of spin orthogonality in the magnetic structure.[17] According to free energy calculations, opposite signs between the Nd^{3+}-Nd^{3+} and the Cu^{2+}-Cu^{2+} interactions would be responsible for this symmetry lowering.[18] More recently, acoustic measurements at lower frequencies performed on polycrystalline NCCO bars revealed a softening of the Young's modulus below 20 K.[19] Because Ce doping shifts this anomaly to lower temperatures, the authors concluded that it could not be related to the minimum observed in C_{66}(T) in the ultrasonic experiments performed on single crystals.[16,17] The authors interpreted rather this anomaly in terms of a paraelastic contribution from the relaxation of Nd^{3+} $4f$ electrons between the levels of the ground state doublet split by the interaction with the antiferromagnetically ordered Cu^{2+} lattice; the value of the splitting was then found to be in agreement with inelastic neutron scattering,[20] infrared transmission,[21] and specific heat experiments.[22]

In this paper, we further investigate the magnetic properties of Nd_2CuO_4 at low temperature with the ultrasonic technique. Beyond the confirmation of previous data, we have characterized more thoroughly the low temperature anomalies observed around 5 K by performing experiments as a function of frequency and magnetic field. We focused our study on the C_{66} elastic constant, which exhibits the strongest anomalies at low temperatures, and on the corresponding attenuation variations. Neither a spontaneous ferrroelastic transition[16,17] nor a paraelastic contribution[19] can explain our results; we rather propose to relate these anomalies to growing magnetic domains due to the frustration of the Nd^{3+} magnetic subsystem arising from the competition between Nd^{3+}-Cu^{2+} and Nd^{3+}-Nd^{3+} interactions.

II. EXPERIMENT

Preliminary ultrasonic measurements on our large Nd_2CuO_4 samples have indicated that the strong ultrasonic attenuation below 10 K, which increases and distorts the signal as the frequency is enhanced, does not allow a detailed study as a function of frequency. In order to reduce the attenuation significantly and avoid such complications, we chose a Nd_2CuO_4 single crystal (l_a=0.54 mm; l_b=2.37 mm; l_c=0.47 mm) grown by the flux method having a short propagation length l_a. Usually, samples are measured using an ultrasonic interferometer working either in the transmission or the reflection modes. However, samples with a short propagation length cannot be studied with the standard multiple echo technique since the acoustic echoes overlap and cannot be separated from the electric pulse. For this reason, a CaF_2 delay line is introduced between the sample and the receiving transducer to enable measurements of small samples in the transmission mode, for the first transmitted pulse only.[23,24] Using a frequency feedback, the phase shift of the first transmitted pulse is maintained constant with respect to a reference signal. Both the amplitude and the frequency variations of that pulse are measured. This modified technique generally yield highly reliable results for the velocity variations since it is based on a null signal detection. The amplitude of the transmitted echo can, however, be weakly polluted by interference effects originating from multiple reflections inside the crystal, which overlap with various relative phases. The use of $LiNbO_3$ transducers allows us to generate and detect, at 24 MHz and corresponding odd overtones, transverse waves propagating along the *a* axis and polarized along the *b* axis (C_{66}). While the absolute attenuation and elastic constant values are not accessible, the variations of attenuation $\alpha(T)-\alpha(40\text{ K})$ and the relative variations of the elastic constant $[C_{66}(T)\text{-}C_{66}(40\text{ K})]/C_{66}(40\text{ K})$ are obtained with high accuracy. A magnetic field (0–10 T) has been applied along various orientations. Both the variation of the sound attenuation and the relative variation of the elastic modulus C_{66} were obtained in the 1.7–80 K temperature range.

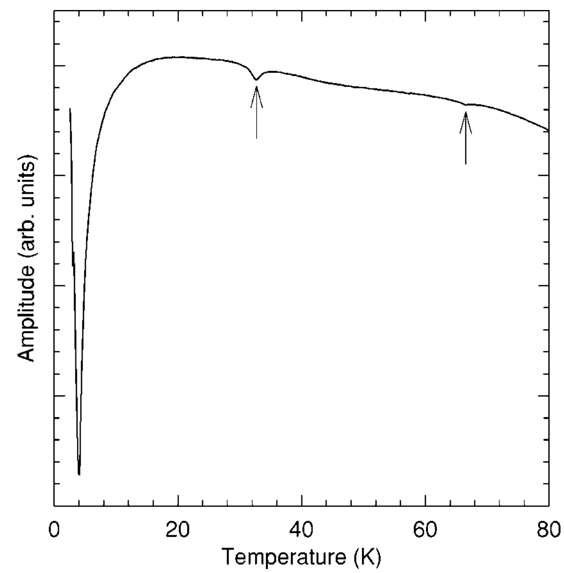

FIG. 1. Amplitude of the first transmitted pulse at 208 MHz. Arrows correspond to Cu^{2+} spin reorientations.

III. RESULTS AND DISCUSSION

Figure 1 shows the amplitude of the first transmitted pulse at 208 MHz, in the 2–80 K temperature range. As the temperature is decreased from 80 K, three dips are observed around 67, 33, and 4 K, respectively. The dip intensities are enhanced as the temperature is decreased similarly to the Nd^{3+} magnetic susceptibility,[14,25] suggesting a magnetoelastic coupling involving directly the Nd^{3+} magnetic moments rather than the Cu^{2+} ones. Furthermore, the Cu^{2+} Néel phase transition has been detected neither in our measurements nor in previous ultrasonic studies,[16,17] indicating a weak coupling of the Cu^{2+} sublattice with the acoustic phonons. The first dip observed at 67 K, which is revealed using the ultrasonic technique, is associated with phase I→phase II transition. According to the enhancement of the attenuation with frequency, the corresponding anomaly could not be detected below 208 MHz. Also, no anomaly could be identified out of the noise on the relative variations of the elastic constant C_{66} around 67 K. The second dip is associated with the phase II→phase III magnetic transition, as reported elsewhere.[16] The anomaly around 4 K, which is much stronger than the others, is the main subject of this paper. It is also worth noting that no hysteresis associated with the measurements has been observed.

The attenuation variations $\Delta\alpha(T)$ and the relative variations of the elastic constant C_{66} due to the phase II→phase III magnetic transition, are given in Fig. 2. For sake of clarity, the $\Delta C_{66}(T)/C_{66}$ curves obtained at various frequencies

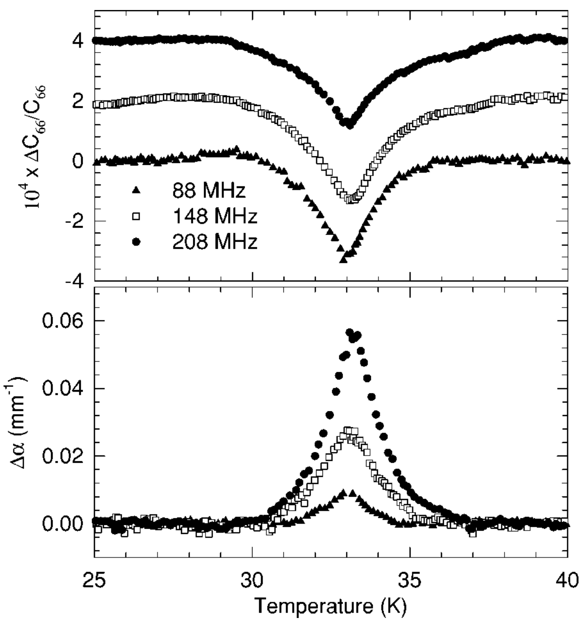

FIG. 2. Relative variation of the elastic constant C_{66} (top panel) and variation of attenuation (bottom panel) due to the phase II→phase III magnetic transition. The $\Delta C_{66}/C_{66}$ curves have been shifted for sake of clarity.

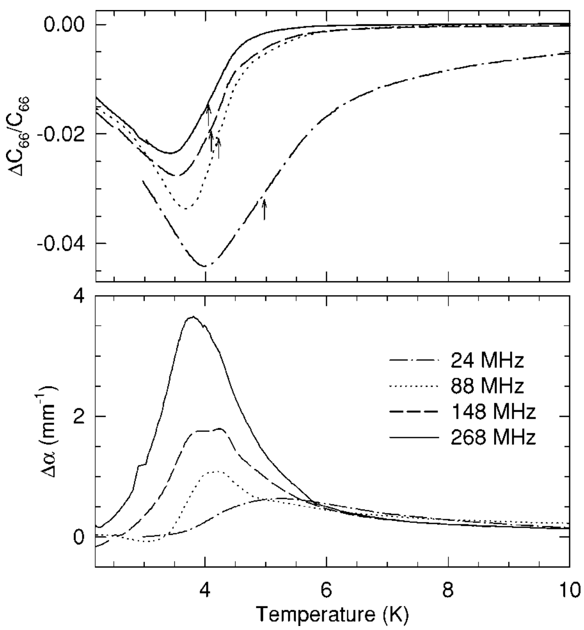

FIG. 3. Relative variation of the elastic constant C_{66} (top panel) and variation of the attenuation (bottom panel) at various frequencies, measured using a transverse wave propagating along the *a* axis and polarized along the *b* axis. The arrows correspond to the critical temperature T_a (see the text).

have been separated upwards from each other. Both $\Delta\alpha(T)$ and $\Delta C_{66}(T)/C_{66}$ exhibit a symmetric anomaly around 33 K. The shape of the anomalies supports the hypothesis of a spin reorientation. While a peak of attenuation is observed during the rotation of the Nd^{3+} and the Cu^{2+} spins, a small softening occurs in $\Delta C_{66}(T)/C_{66}$. Furthermore, the variations in the attenuation and in the elastic constant C_{66} are the same below and above the transition. This indicates that the magnetic structure has not been significantly modified by the spin reorientation. While $\Delta C_{66}(T)/C_{66}$ is not affected by the used ν frequency, the amplitude of the $\Delta\alpha(T)$ anomaly varies with a ν^2 frequency dependence. Moreover, the temperature at which the anomaly occurs is not frequency dependent, as it is expected at a phase transition.

The situation is more complicated in the case of the anomaly around 4 K, as illustrated in Fig. 3, which shows $\Delta C_{66}/C_{66}$ and $\Delta\alpha$ at various frequencies. Both the $\Delta\alpha$ and the $\Delta C_{66}/C_{66}$ anomalies around 4 K are stronger than those detected at 33 K by two orders of magnitude, due to a much stronger elastic coupling compatible with the Nd^{3+} magnetic moments reinforcement at low temperature. As for the transition at 33 K, a peak of attenuation is accompanied by a softening of the C_{66} elastic constant. However, the anomalies around 4 K differ from those at 33 K in their amplitude, shape and frequency dependence.

Contrary to the phase II→phase III transition, the $\Delta\alpha$ and $\Delta C_{66}/C_{66}$ anomalies are not symmetric and their temperature profiles differ below and above the transition. The small wiggles observed on the attenuation data are extrinsic effects likely originating from overlapping echoes: they are not observed systematically at all frequencies and they have no counterparts on the variations of the elastic constant. These wiggles are observed only in the temperature range where the variations of the elastic constant $C_{66}(T)$ vary rapidly, leading to strong modulations of the relative phases between the overlapping echoes and consequently to modulations of the measured attenuation amplitude. Interestingly, the temperature for which the attenuation is maximum does not correspond to the minimum of $\Delta C_{66}/C_{66}$, but is rather close to the maximum of its temperature derivative, indicated in Fig. 3 by upward arrows. Also noticeable is the frequency dependence of the 4 K anomalies. As shown in Fig. 3, the $\Delta C_{66}/C_{66}$ and the $\Delta\alpha$ anomalies are shifted towards lower temperatures as the frequency is enhanced. This shift is accompanied respectively by a decrease and an increase in the amplitude of the $\Delta C_{66}/C_{66}$ and the $\Delta\alpha$ anomalies. The temperature profiles of the anomalies sharpen as frequency increases and the reinforcement of the $\Delta\alpha$ anomaly amplitude follows a ν dependence rather than a ν^2 dependence. Such a frequency dependence of $\Delta\alpha$ and $\Delta C_{66}/C_{66}$ cannot be attributed to a phase transition. This situation suggests that the anomalies are related either to a resonance or a relaxation phenomenon characterized by coefficients varying with temperature. However, the relaxation model would not produce a peak in $\Delta C_{66}/C_{66}$ unless the relaxation time $\tau(T)$ does not vary monotonically with temperature. For this reason, we argue that the anomalies shown in Fig. 3 are more compatible with a resonance phenomenon. For example, such a model has already been introduced in order to describe the damping of ultrasonic waves due to dislocations.[26] Hence, $\Delta\alpha$ and $\Delta C_{66}/C_{66}$ could be related, respectively, to the dissipative and the elastic terms of a forced harmonic oscillator response to a fixed excitation frequency, when the parameters which characterize the resonance phenomenon vary with temperature.

Since the 4 K anomalies are observed only in $Nd_{2-x}Ce_xCuO_4$, they should be related to the Nd^{3+} ion mo-

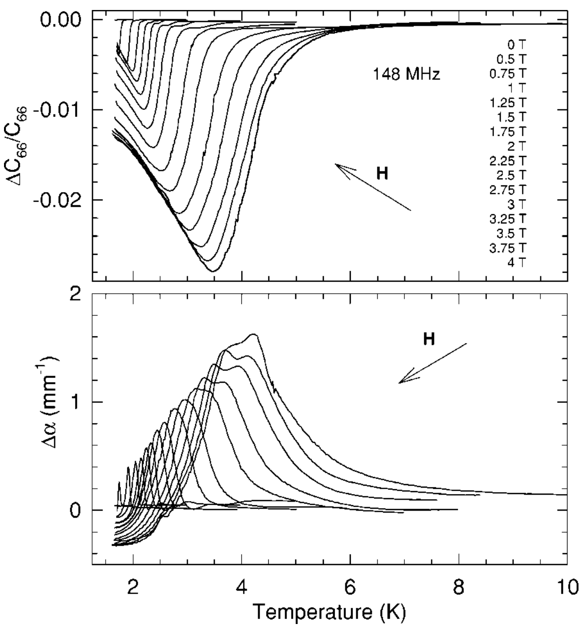

FIG. 4. Relative variation of the elastic constant C_{66} (top panel) and variation of the attenuation (bottom panel) at 148 MHz under magnetic field. The field is applied along the *a* axis (propagation direction).

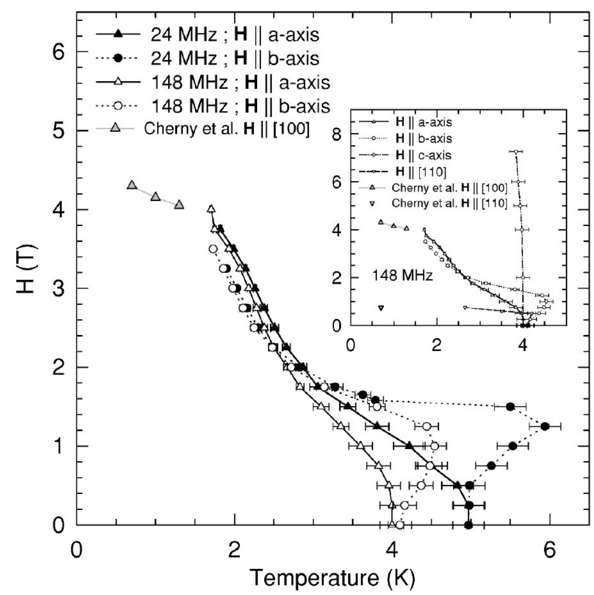

FIG. 5. Magnetic field dependence of the low temperature anomaly critical temperature T_a, at 24 and 148 MHz, for field applied along the *a* axis and along the *b* axis. The magnetic field dependence of T_a at 148 MHz for various field orientations are shown in the inset. The gray symbols refer to magnetization data from Cherny *et al.* (see Ref. 11). Lines are guides for the eye.

ments. In order to confirm that these anomalies are the consequences of their magnetic properties, measurements have been performed under magnetic field. The low temperature magnetic field dependences of $\Delta C_{66}/C_{66}$ and $\Delta\alpha$ at 148 MHz, which are illustrated respectively in the top and bottom panels of Fig. 4 for a field applied along the *a* axis (propagation direction), confirm that the anomalies have a magnetic origin. The influence of the magnetic field on the 4 K anomalies is similar to the one produced by a frequency increase: while these anomalies are shifted towards lower temperatures, they get sharper as the magnetic field is enhanced. However, both amplitudes of the $\Delta C_{66}/C_{66}$ and the $\Delta\alpha$ anomalies decrease under magnetic field and no anomaly is observed above 4 T in our temperature range. Furthermore, the temperature profiles of $\Delta C_{66}/C_{66}$ become very different above and below the minimum of the $\Delta C_{66}/C_{66}$ anomaly. While the profile of $\Delta C_{66}/C_{66}$ above that minimum is very abrupt above 2 T, it is not significantly affected by the applied field below 2 T. Moreover, all the $\Delta C_{66}/C_{66}$ curves corresponding to $H<2$ T reach the same curve below the minimum of the anomaly. Above that field, the low temperature values of $\Delta C_{66}/C_{66}$ are no longer in coincidence.

As mentioned earlier, the $\Delta C_{66}/C_{66}$ and the $\Delta\alpha$ anomalies around 4 K can be interpreted as the elastic and the dissipative terms of a forced harmonic oscillator response, respectively. The two parameters which characterize the resonance phenomenon are the center temperature T_a (defined as the maximum temperature derivative of $\Delta C_{66}/C_{66}$) and the full width ΔT_a. On the different plots shown in Figs. 3 and 4, only T_a values could be measured with high precision at different frequencies and magnetic field values. These T_a data are reported in Fig. 5 as a function of magnetic field at 24 and 148 MHz frequencies. Similar data at other frequencies are not included for the sake of clarity.

Even though the *a* and the *b* axes are magnetically equivalent, we have included in Fig. 5 data obtained with the field applied along these two axes. We recall that we use transverse waves propagating along the *a* axis and polarized along the *b* axis to obtain C_{66}; different field orientations imply different orientations of the Nd^{3+} moments relative to the polarization vector $\hat{\mathbf{e}}$. For $\mathbf{H}\perp\hat{\mathbf{e}}$ ($\mathbf{H}\parallel a$ axis, black and white upward triangles), the characteristic temperature T_a decreases with increasing field. If the values of T_a are quite different in zero field for the two frequencies, they merge as a single line above 2 T: this behavior has been verified at all frequencies. For $\mathbf{H}\parallel\hat{\mathbf{e}}$, ($\mathbf{H}\parallel b$ axis, black and white circles), T_a rather increases with field for $H\lesssim 1.5$ T; it then decreases rapidly for higher field values before reaching the universal *H-T* line observed for the other field orientation. These measurements confirm that the low temperature $\Delta C_{66}/C_{66}$ and $\Delta\alpha$ anomalies cannot be related to a magnetic phase transition, in contradiction with previous studies using only one frequency.[16–18]

While the Nd^{3+} magnetic subsystem is involved in the resonance phenomenon that occurs around 4 K, a strong Nd^{3+}-Cu^{2+} coupling remains, as indicated by data obtained with various magnetic field orientations. The results at 148 MHz are reported in the inset of Fig. 5. Similarly to the anomalies at 33 and 67 K, which are related to the Cu^{2+} spins coupled to the Nd^{3+} moments, the low temperature anomalies are not significantly affected by a magnetic field as high as 7.25 T applied along the *c* axis. Only a small shift of T_a for $\mathbf{H}\parallel c$ axis is observed, possibly due to a small canting of the Nd^{3+} moments out of the *ab* planes. While the 4 K anomaly disappears above a field of 4 T applied along the *a* and *b* axes, a more drastic effect is observed when the field is oriented along the [110] direction. In such a configuration, T_a

decreases rapidly above 0.5 T, and persists up to 0.75 T. These results are consistent with magnetization data on Nd_2CuO_4, which are most sensitive to the Nd^{3+} magnetic moments. Considering that the Nd^{3+} spin orientation is driven by the Cu^{2+} magnetic sublattice, a magnetization study has reported critical fields of $H_{c1}=4.4$ T and $H_{c2}=0.75$ T associated with the transition from a noncollinear to a collinear magnetic structure at 0.7 K, for magnetic fields applied along the [100] and the [110] directions, respectively.[11] However, these transitions are detected only at low temperature and are not observed above 2.2 K. The values of T_a obtained from magnetization have been included in Fig. 5 (gray triangles) and we observe that our universal *H-T* line extrapolates towards these magnetization data.

Now we may ask the question: what is the origin of the 4 K anomaly observed in $Nd_{2-x}Ce_xCuO_4$ crystals? On the one hand, a paraelastic contribution due to the splitting of the Nd^{3+} Kramers ground state doublet as suggested by Cordero *et al.*[19] cannot explain the frequency and magnetic field dependences observed in our ultrasonic experiment. Indeed, a paraelastic contribution should decrease in amplitude with increasing frequency without any shift in temperature; moreover, since the ground state splitting should increase with magnetic field, the anomalies would then move to higher temperatures, as the Schottky anomaly in the specific heat measurements.[22] The frequency and magnetic field trends reported here are opposite to these observations. If a paraelastic contribution is still possible in this system, it is not detected in the megahertz frequency range. On the other hand, although our data are fully consistent with previous ultrasonic studies, they cannot be explained by a spontaneous ferroelastic phase transition.[16–18] The frequency and the magnetic field orientation (*a* axis versus *b* axis) dependences of these anomalies obtained in the present study invalidate this hypothesis since the energy of the probe used is much lower than the typical energies involved. These dependences are also incompatible with a possible resonance of the ultrasonic wave with the gap observed below 4.5 T in an acoustic magnon branch:[14] the gap energy being two to three orders of magnitude larger than the energies associated with the used ultrasonic frequencies ($1\ \mathrm{meV} \approx 2.4\times10^5$ MHz). Besides, thermal conductivity measurements show similar results in Nd_2CuO_4 and Pr_2CuO_4[14] while the ultrasonic anomaly around 4 K is observed only in $Nd_{2-x}Ce_xCuO_4$ crystals.

Below 2 T, the experimental data of Fig. 5 rather suggest the presence of domains, which resonate with the ultrasonic waves. Hence, these domains should involve either a characteristic length or a characteristic time. Using the absolute sound velocity of a transverse wave propagating along the *a* axis and polarized along the *b* axis, which is 4.22×10^3m/s,[27] the corresponding length or correlation length that would be probed in our experiment ranges between 15 and 175 μm. On the other hand, the corresponding time or correlation time that would be probed ranges between 4 and 42 ps. These domains could also be related to a Nd^{3+} spin glass phase.

A likely candidate to explain the development of these magnetic domains is the competition between the Nd^{3+}-Cu^{2+} exchange interaction, which is responsible for the polarization of the Nd^{3+} sublattice at temperatures as high as 140 K,[7] and the Nd^{3+}-Nd^{3+} interaction which is enhanced significantly at low temperature. In Fig. 5, the discrepancy observed between the frequency dependent regime data and the extrapolation to lower field of the *H-T* universal line obtained above 2 T suggests that the magnetic structure is frustrated. Hence, as the temperature is lowered, the magnetic moments of the Nd^{3+} Kramers ions increase rapidly, as indicated by the in-plane Nd_2CuO_4 susceptibility.[13,14,25] Consequently, the frustrated magnetic structure of the Nd^{3+} subsystem gets unstable. Knigavko *et al.* have suggested that the Nd^{3+}-Nd^{3+} and the Cu^{2+}-Cu^{2+} interactions have opposite signs.[18] This idea has also been pointed out in an inelastic neutron scattering study.[28] Consequently, the orientation of the Nd^{3+} ions, as imposed by the Cu^{2+} subsystem *via* the Nd^{3+}-Cu^{2+} anisotropic exchange interaction is not the one adopted in the absence of the Cu^{2+} subsystem.

The frustration of the Nd^{3+} magnetic subsystem should lead to the formation of local magnetic domains where the Nd^{3+} and Cu^{2+} magnetic moments are not parallel. Hence, there are some indications that the magnetic structure of Nd_2CuO_4 at low temperature has a lower symmetry than the usually reported one. While the supposed *mmm* symmetry of the Nd_2CuO_4 magnetic structure does not allow any magnetoelectric effect, a non-negligible electric polarization along the [100] direction is observed at 4.2 K as an external magnetic field is applied along the [010] direction.[29] Moreover, a splitting along the AM direction of the Nd^{3+} spin wave dispersion[30] has been attributed to a tilting of the Nd^{3+} spins with respect to the Cu^{2+} sublattice, which is estimated to be less than 1°.[31]

The *H-T* line above a magnetic field of 2 T applied along the Cu-O bonds could be associated with a field-induced transition of the Nd^{3+} spins from a noncollinear to a collinear magnetic structure. While the field-induced transition of the Cu^{2+} subsystem is observed up to 18 K in thermal conductivity without significant variation of the critical field $H_{c1}=4.5$ T,[14] the magnetization of Nd_2CuO_4, directly sensitive to the Nd^{3+} magnetic moments, shows small variations of that critical field which extrapolate to the *H-T* line measured with the ultrasonic technique. Nevertheless, these magnetization data have been unambiguously attributed to a transition from a noncollinear to a collinear magnetic structure.[11] Hence, the noncollinear magnetic structure of the Nd^{3+} subsystem at low temperature transits to a collinear configuration at a critical field lower than the corresponding field-induced transition of the Cu^{2+} spins, in contrast to the common belief.[11] On the other hand, the magnetic anisotropy between the *a* and *b* axes, as well as the frequency dependence of the *H-T* line below 2 T, suggest that the Nd^{3+} noncollinear→collinear magnetic transition is not complete in this range, but is rather characterized by magnetic domains which indicate a beginning of Nd^{3+} ordering. Hence, no long range self-ordering of the Nd^{3+} spins is observed below 2 T.

Even though the characterization of the resonance phenomenon due to the presence of these magnetic domains would need a better understanding of their magnetic structure, which cannot be determined with only ultrasonic measurements, we can adequately assume that the damping coefficient and the resonance frequency of the resonance

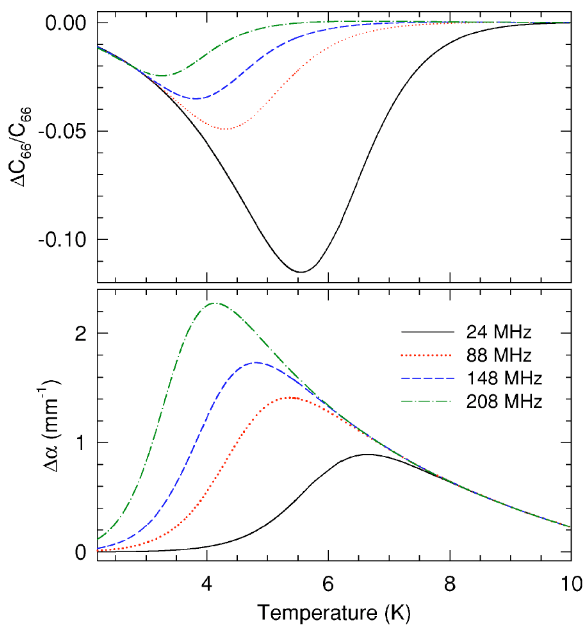

FIG. 6. (Color online). Simulated relative variation of the elastic constant C_{66} (top panel) and variation of the attenuation (bottom panel) obtained at various frequencies using the resonance model described in the text.

phenomenon are somehow related to the Nd^{3+} uniform magnetic susceptibility $\chi(\mathbf{q}=\mathbf{0},T)$, which increases rapidly at low temperature. In the relaxation model, frequently used for magnetic domains,[32,33] the damping coefficient is supposed to be inversely proportional to $\chi(\mathbf{q}=\mathbf{0},T)$ and such a dependence will also apply here. As for the resonance frequency, we expect that it will increase with the domains magnetization, and thus be proportional to $\chi(\mathbf{q}=\mathbf{0},T)$. Such temperature dependent parameters are sufficient to reproduce qualitatively the attenuation and the elastic constant C_{66}(T) observed experimentally. According to Ref. 26, ΔC_{66}(T) and $\Delta\alpha$(T) can be described, in the case of a resonant phenomenon, by

$$\Delta C_{66}(T) = A\frac{\nu^2 - \nu_0^2(T)}{[\nu^2 - \nu_0^2(T)]^2 + \Gamma^2(T)\nu^2}, \quad (1)$$

$$\Delta\alpha(T) = B\nu\frac{\Gamma(T)\nu}{[\nu^2 - \nu_0^2(T)]^2 + \Gamma^2(T)\nu^2}, \quad (2)$$

where A and B are constants. Using the strongly temperature dependent parameters $\Gamma(\mathrm{T}) \propto 1/\chi(\mathbf{q}=\mathbf{0},T)$ and $\nu_0 \propto \chi(\mathbf{q}=\mathbf{0},T)$, where the uniform paramagnetic susceptibility $\chi(\mathbf{q}=\mathbf{0},T)$ can be obtained from the the crystal-field parameters given in Ref. 34, simulations of ΔC_{66}(T) and $\Delta\alpha$(T) at various frequencies have been performed and reported in Fig. 6. Even though the Nd^{3+}-Nd^{3+} and Nd^{3+}-Cu^{2+} interactions have not been included in the calculation of $\chi(\mathbf{q}=\mathbf{0},T)$, these simulations reproduce rather very well both the shapes and the frequency dependence of the experimental data shown in Fig. 3, especially the displacement with frequency, confirming that the ultrasonic anomalies found around 4 K on single crystals are described by a resonant phenomenon involving strongly temperature dependent parameters, rather than by a relaxation process. A quantitative comparison will be only possible using an adequate model including the Nd^{3+}-Nd^{3+} and Nd^{3+}-Cu^{2+} interactions, which is outside the scope of the present study.

IV. CONCLUSION

In this study, we have shown that the ultrasonic technique is a powerful tool to characterize the magnetic structure of Nd_2CuO_4. In addition to an anomaly related to the phase II→phase III transition, an anomaly related to the phase I→phase II transition is observed using this technique. Moreover, the use of small samples has allowed a frequency study of the ultrasonic anomalies previously reported around 4 K on both the ultrasonic attenuation and the C_{66} elastic modulus. These anomalies have been found to be field orientation and frequency dependent, preventing their association with a phase transition, in contrast to previous studies using only one ultrasonic frequency. This behavior is also incompatible with the resonance of an ultrasonic wave with an energy gap in an acoustic magnon branch.[14] Owing to the anomaly profiles, which cannot be reproduced by any relaxation model using a relaxation time varying monotonically with temperature, we propose that these anomalies are associated with a resonance phenomenon which is somehow related to local magnetic domains due to the frustration of the Nd^{3+} magnetic subsystem arising from the competition between the Nd^{3+}-Cu^{2+} and the Nd^{3+}-Nd^{3+} interactions. The latter increase as the Nd^{3+} low temperature magnetic moments. Finally, we have shown that the Nd^{3+} magnetic moments cannot order by themselves without a magnetic field of 2 T applied along the Cu-O bonds. Above that critical field, the *H-T* line obtained is associated with a transition of the Nd^{3+} spins from a noncollinear to a collinear magnetic structure, which occurs at a lower critical field than the one required for the Cu^{2+} spins.

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*Electronic address: pierre.richard@bc.edu

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