Vortex matter in the type-II superconductor \(\text{La}_3\text{Ni}_2\text{B}_2\text{N}_3-x\) in the light of NMR

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Abstract. A quantitative analysis of spin-echo NMR measurements of the local field distribution and the spin–spin relaxation time \(T_2\) in the field-cooled superconducting state of the type-II superconductor \(\text{La}_3\text{Ni}_2\text{B}_2\text{N}_3-x\) shows evidence for two fluctuation modes of the weakly pinned vortex matter dominating \(T_2\) at different temperatures: long wavelength diffusive modes in the vortex solid at low temperature, and plastic deformation of the viscous liquid close to the critical temperature \(T_c\), with a broad crossover region in between where a more complex theory is required. We determine the melting temperature \(T_m = 11.5\) K of the vortex matter as the temperature where the characteristic time of the plastic deformation slows down to the time scale set by the spin-echo experiment. This is preferred to the conventional estimates from a Lindemann criterion for the melting vortex solid since the quantitative description of \(T_2\) near \(T_m\) by the viscous vortex liquid appears to be more consistent. It indicates that the plastic deformation mode might be important for \(T_2\) in the mixed state of other type-II superconductors in the weak pinning regime near \(T_c\) as well.
1. Introduction

The properties of flux lines in type-II superconductors pose challenging problems for theory as well as for technical applications. In particular with the advent of high-$T_c$ cuprate superconductors, a very complex field–temperature ($H$–$T$) phase diagram in the superconducting phase became evident, which depends strongly on intrinsic material parameters as well as on the properties of the defect structures shaping the pinning potential [1, 2]. For technical applications, the extended regimes of flux flow and flux creep in critical states with macroscopic shielding or transport currents $j_m$ near critical currents $j_c$ are crucial. A most intriguing aspect of the $H$–$T$ phase diagram is the possibility of a liquid–solid phase transition of the vortex matter [3–6]. Depending on the parameters of the material and the pinning potential, the transition can occur well separated from the transition between the superconducting and normal state at $B_{c2}(T)$.

NMR has frequently been used to detect thermal fluctuations of the flux lines or fluctuations from current-driven flux flow through corresponding fluctuations of the local field (see e.g. [7–13]). In this work we present evidence from NMR and magnetization measurements that La$_3$Ni$_2$B$_2$N$_{3-x}$ represents an example from the NiB-superconductor family where the complex $H$–$T$ phase diagram can be studied. The Abrikosov–Gorkov parameter $\kappa$ is smaller than for the high-$T_c$ cuprates but still large enough to expect a liquid phase of the flux line lattice (FLL) down to temperatures significantly below the critical temperature at the corresponding field. We use the resonance of $^{11}$B as an example for a detailed discussion of the fingerprint that the FLL in thermal equilibrium leaves in the NMR linewidth and spin–spin relaxation rate $1/T_2$.

We propose treating the vortex matter and its thermal fluctuations close to $B_{c2}(T)$ as a viscous liquid instead of using, as is conventionally done, the theory of elasticity.

2. Sample characterization

La$_3$Ni$_2$B$_2$N$_{3-x}$ crystallizes in the space group $I4/mmm$ (no. 139) with one formula unit per primitive unit cell [14]. The structure may be visualized as NiB double layers separated by three layers of LaN [15]. Polycrystalline samples were grown following the procedure described elsewhere [16, 17] and ground into coarse powders (grain diameters 0.1–1.0 mm). We did not succeed in growing stoichiometric samples ($x = 0$). Our attempts to influence the carrier concentration by adjusting the number of nitrogen vacancies in the central LaN layer...
revealed a rather narrow range ($x = 0.06–0.1$) for the variation of the nitrogen stoichiometry in La$_3$Ni$_2$B$_2$N$_{3-x}$ [18]. The shape of the transition to the superconducting state observed in specific heat, susceptibility, as well as the NMR spin–spin relaxation time in the superconducting state described below is very sensitive to the thermal treatment of the samples, indicating a strong influence of the microstructure formed by the defects. We note, however, that the NiB planes are well shielded from the defects in the central LaN planes since neither the $^{11}$B NMR spectra nor their relaxation in the normal state is affected [15].

Figure 1 shows the characteristics of the $H$–$T$-phase diagram discussed in the context of our NMR results below for the sample with optimum thermal treatment. The superconducting transition temperature in zero field depends on the nitrogen vacancy concentration and is for this sample $T_c = 13.7$ K. From specific heat measurements the upper critical field at zero temperature is $B_{c2}(0) = 9.3$ T, the penetration depth is $\lambda_{(0)} = 230$ nm and the Abrikosov–Gorkov parameter is $\kappa = \lambda/\xi = 41$. Up to now we were unable to prepare suitable single crystals for measurements of the anisotropy of these parameters. For this work we approximate the critical line by $B_{c2}(T) = B_{c2(T=0)}(1 - T/T_c)^{2}$, that is within the Ginzburg–Landau theory underlying the theory of elasticity of the FLL discussed below. The irreversibility line was determined by measuring the time dependence of the magnetization at fixed temperatures following finite field steps (0.1 T per step). The observed time dependence is approximately logarithmic, as expected for flux creep. The irreversibility line in the figure indicates where the time constant becomes smaller than the time resolution (1 s). This shows that our field-cooled NMR experiment at $B_0 = 1.293$ T probes the state of the FLL above the irreversibility line at all temperatures. The regime below the irreversibility line is difficult to investigate by NMR since frequencies below 5 MHz would be encountered.
Figure 2. Upper panel: local field distribution at $^{11}$B derived from NMR spin-echo spectra (central transition) at various temperatures in an applied field $B_0 = 1.293$ T. The red lines are fits by one or two Gaussians. The lower panel shows the full linewidth (twice the second moment of the local field distributions calculated directly from the data, $\Delta$), and the field difference between the two unresolved lines at temperatures below 9.5 K ($\nabla$). The green dots show the temperature dependence of the difference between local and applied field relative to the applied one calculated from positions of the Gaussians (Knight shift).

3. Static inhomogeneous line broadening from the flux line lattice

Figure 2 shows in the upper panel the Fourier transform of the spin echo of the $^{11}$B central line at $B_0 = 1.293$ T in a field cooling experiment in the normal state (at $T = 16$ K), and at several temperatures in the superconducting state for the sample with the narrowest transition region. The spectra are normalized and the frequency axis has been divided by the gyromagnetic ratio $\gamma = 13.660$ MHz T$^{-1}$ of $^{11}$B to represent the distribution of the local field $B_l$. Given the very close to Gaussian line shapes we determined peak positions from Gaussian fits to the data, while overall linewidths were evaluated directly from the data as the square root of second moments. The lower panel shows the increase of the linewidths with respect to the normal state linewidth, and the temperature dependence of the line position. The latter shows the onset of a homogeneous diamagnetic response at $T_c = 12.7$ K in this field (vertical line), in accord with the phase diagram (figure 1) and the spin–spin relaxation discussed below. Note that the difference between the mean internal field, given by the flux line density $n$ times the flux quantum $\Phi_0$, and the applied field is always small compared to the applied field.
The nuclear spins are subject to local fields depending on the position of the nuclei with respect to the flux lines. For ideal hexagonal or square Abrikosov lattices a single peak in the local field distribution has been calculated and is well established experimentally \[7, 19, 20\]. The highest local fields correspond to the kernel of the flux lines where a field in the order of the lower critical field \(B_{c1}\) penetrates on a diameter of approximately the coherence length \(\xi(T)\) \[1\]. The surrounding superconductor is shielded on the length scale of the penetration depth \(\lambda(T)\) with minimum field at the center of the FLL unit cell. The field with maximum probability corresponds to the saddle points between flux lines. This peak is broadened if the flux lines are allowed to bend and adjust their positions in the random pinning potential. A double peak structure of the local field distribution indicates the presence of two different characteristic vortex distances or shielding length scales. Strongly anisotropic superconductivity can result in sheared Abrikosov lattices and is a possible intrinsic origin for this \[20, 21\]. In La\(_3\)Ni\(_2\)B\(_2\)N\(_3\)\(_{−x}\), however, the width of the superconducting specific heat anomaly in applied fields shows no evidence for any significant anisotropy \[22\] and justifies our use of the isotropic theory throughout this work.

In La\(_3\)Ni\(_2\)B\(_2\)N\(_3\)\(_{−x}\) the linewidth in the field-cooled mixed state stays constant down to approx. 11.3 K, in a regime where the resistivity of the sample drops significantly with temperature. A similar observation in high-\(T_c\) cuprates has been attributed to rapidly moving flux lines hidden by motional narrowing \[12\]. It indicates that the motion of the flux lines in the vortex liquid at temperatures near \(T_c\) is fast enough to average the local field within the time of a nuclear spin precession. At lower temperature the time scale of these motions increases and the field inhomogeneities freeze into the observed line broadening. The spectra in figure 2 develop an unresolved double peak structure below approx. 9.5 K. In view of the rather large inhomogeneous Gaussian linewidth even above \(T_c\), and of the unknown origin of the splitting, we do not attempt to fit the structure by a complex vortex lattice structure. For the discussion below, only the magnitude of the local field variation is important. The red line shows that the size of the broadening is fully consistent with the second moment for the field distribution of a regular hexagonal FLL, \(\sqrt{\Delta b^2} = 0.061\Phi_0/\lambda^2\), calculated by Brandt \[23\]. The numerical prefactor depends only slightly on the geometry of the vortex lattice. We inserted the flux quantum \(\Phi_0\) and the Ginzburg–Landau penetration depth \(\lambda(T) = \lambda_0/\sqrt{1-T/T_c}\) with \(\lambda_0 = 200\) nm. The fact that this value is within the errors determined in independent specific heat measurements \[17\] justifies our use of this approximation for the static linewidth to estimate the amplitude of the field fluctuations induced by moving vortices in the next section.

4. Spin–spin relaxation induced by vortex fluctuations

In figure 3 we compare the temperature dependence of \(1/T_2\) in field cooling experiments of three different samples. The samples have increasingly inhomogeneous transitions to the superconducting state in specific heat and susceptibility measurements (not shown) due to different thermal treatments. All observed spin-echo decays were single exponential and with equal time constants for the two unresolved peaks at low temperature. Above \(T_c\) (and below 25 K), \(1/T_{2nm} = 1/700\) \(\mu s^{-1}\) is nearly temperature independent and similar for all three samples. The rates \(1/T_2\) shown for the more inhomogeneous samples are increased by 10 and 15\%, respectively. For the most homogeneous sample, data from Hahn-echo and from Carr–Purcell multipulse sequences are superimposed. The latter have been multiplied by a factor of 1.7 to
show that the decay time in the Carr–Purcell pulse train is longer, as expected e.g. in the presence of spin diffusion [24], but that this factor is independent of temperature across $T_c$.

Second-order time-dependent perturbation theory for the nuclear spin Hamiltonian $H_z = \gamma I_z B_l$ of independent nuclear spins $I_z$ with respect to a small fluctuating field with vector $b_f(t)$ shows that [25]

$$\frac{1}{T_2} = \sum_\alpha J_\alpha(0) + \sum_\beta J_\beta(\omega_l).$$

(1)

The spectral densities $J_\alpha(\omega)$ are the Fourier transforms of the time-correlation functions $g_\alpha(\tau) = \gamma^2 b_{\alpha,\alpha}(t) b_{\alpha,\alpha}(t + \tau)$ of component $\alpha = x, y, z$ of the perturbing field, taken at the resonance frequency $\omega_l$ in the static local field for the so-called non-adiabatic contributions $\alpha = x, y,$ and at zero for adiabatic contributions $\alpha = z$. The first sum contains, therefore, only contributions where the perturbation commutes with the static Hamiltonian. The second sum contains the same transverse field fluctuations as those contributing to the nuclear spin–lattice relaxation $1/T_1$.

First we note that the fluctuations responsible for the maximum in $1/T_2$ below $T_c$ are very anisotropic with fields exclusively along the $z$-direction. $1/T_1$ in the normal state just above $T_c$ is a factor of 1500 smaller [15] than the spin–spin relaxation rate $1/T_{2nm} = 1/700 \mu s$ and drops even further below $T_c$ without any evidence for the contributions corresponding to the second sum in equation (1). Therefore, the longitudinal fluctuations completely dominate in $La_3Ni_2B_2N_3-x$, while the second sum is negligible. The first sum contains the electronic contributions from the normal metal, which we assume to be independent of temperature, as well as the contributions due to fluctuations of the flux line positions. The strong field anisotropy of the latter indicates straight flux lines along the $z$-axis. In fact, the transverse contributions have
been investigated in $T_1$ of Tl in high-$T_c$ cuprates where the layered structure favors kinks in the flux lines at pinning centers [9].

An explicit form of $J(\omega)$ which frequently is sufficient to describe the low-frequency limit in equation (1) can be derived within a simple model for the fluctuation process. One assumes that the small local field along $z$ switches randomly with a constant probability between two values $\pm b_t$. The time-correlation function $g_z(\tau)$ becomes an exponential decay from $b_t^2$ at $\tau = 0$ with characteristic time $\tau_c$ [26]; correspondingly the spectral density is a Lorentzian. Equation (1) for this model reads therefore

$$1/T_{2H} = (\gamma b_t)^2 \frac{\tau_c}{1 + (\omega_c \tau_c)^2}. \quad (2)$$

For our spin-echo experiment with pulse distance $d$, we evaluate $J_z(\omega)$ not at $\omega = 0$ but at $\omega_c = \pi/d$. Perturbations varying on a frequency scale $\omega \ll 2\pi/2d$ will be refocused in the spin echo at time $2d$. Experimentally, $d$ cannot be chosen much larger than the decay time $T_{2nm}$; therefore we set in the following $\omega_c \approx \pi/700 \mu s^{-1}$.

As points of caution we note that equation (1) is based on the assumption $\tau_c < d$ and $\tau_1 < \tau_2$. This obviously breaks down if $\tau_c$ diverges near a critical temperature, which might well introduce systematic errors in the analysis near the transition temperature. In addition, other processes clearly may lead to different spectral densities, and even if $J(\omega)$ is a Lorentzian, the sensitivity of the relaxation at an experimental frequency may not be sufficient to filter a particular process and allow its identification.

4.1. The viscous liquid regime

In the liquid phase the flux lines move without preferred positions. We assume, therefore, that the theoretical static field distribution represents the field amplitudes induced at a fixed site by the moving flux lines and approximate the fluctuation amplitude by the full variance of the theoretical field distribution at that temperature (see figure 2):

$$b_t(T) = 0.061 \frac{\Phi_0}{\lambda_0^2}(1 - T/T_c). \quad (3)$$

For any given fluctuation amplitude $b_t$, the relaxation rate as a function of correlation time $\tau_c$ has a maximum value $1/T_{2H}^{\text{max}} = (\gamma b_t)^2/\omega_c$ at $\tau, \omega_c = 1$. We show in figure 3 that the experimental spin–spin relaxation rate is close to this maximum rate (red line) in the whole temperature range 11.5–$T_c$ where we observed a constant static linewidth. This suggests a melting temperature $T_m$ of the vortex lattice near 11.5 K since at that temperature the characteristic time for free vortex motion should become too long for the vortex fluctuations to be effective for $1/T_2$.

We estimate the thermodynamic characteristic time $\tau_{th}$ for the liquid by the time a flux line moves the average flux line distance $a_0 = \sqrt{\Phi_0/B_0} = 40$ nm. This is $a_0/v$, with the flux line velocity $v$ given by the equipartition principle for the kinetic energy of the vortices $E_{\text{kin}} = k_B T/2$ with $E_{\text{kin}} = m_v v^2/2$. We insert the effective vortex mass per unit length [27] $m_v = m_e(3\xi^2 \Delta F/8E_F) \approx 55,000 m_e$, where $E_F = 0.82$ Ryd is the Fermi energy from band structure calculations [15], $\Delta F = 1.05 \times 10^4$ J m$^{-3}$ is the condensation energy into the superconducting state [18] and $m_e$ is the bare electron mass, and find $v(T = 11.5 K) \approx 60$ m s$^{-1}$ for the vortex velocity. This translates to a characteristic time $\tau_{th} \approx 6.6 \times 10^{-10}$ s.
From equation (2) it is immediately clear that fluctuations with this short characteristic time are completely ineffective for spin–spin relaxation. It has been pointed out by Geshkenbein and, in their extensive review, by Blatter et al that in viscous liquids a second time scale $\tau_{pl}$ for plastic deformation exists [2, 28]:

$$\tau_{pl} = \tau_{th} \exp(U_{pl}/T).$$  

(4)

$U_{pl} = U_0(1 - T/T_c)$ with $U_0 = a_0(\Phi_0/(4\pi\lambda_0))^2 = 1550\,\text{K}$ is the energy scale for plastic deformation of the FLL. The barriers are essential to characterize flux flow and thermally activated flux flow behavior, which has been studied extensively in the small current limit of the critical state in high-$T_c$ cuprates [2]. We note that the barrier height $U_0$ is the same that we observed experimentally in the time dependence of magnetization at fields below the irreversibility field (figure 1). The crucial point for NMR is that $\tau_{pl}$ becomes exponentially longer with decreasing temperature and will exceed eventually the characteristic time of the NMR experiment. The crossover should correspond to a maximum in the spin–spin relaxation rate induced by the mode.

Note that this second time scale $\tau_{pl}$ introduces no new fit parameters into equation (2) for $1/T_c$ as long as in a weak pinning limit only the intrinsic energy $U_0$ is considered. The dotted red line results with $\lambda_0 = 200\,\text{nm}$, $T_c = 12.7\,\text{K}$ and $\tau_{th} = 6.6 \times 10^{-10}\,\text{s}$ discussed above, and from the assumption that the plastic deformation shifts the flux lines across distances larger than $a_0$, that is the fluctuating field amplitude is given by equation (3). The narrow temperature window of only $\approx 0.2\,\text{K}$ width where this plastic motion contributes is the result of the steep $\exp(1/T)$ temperature dependence of $\tau_{pl}$. No strong temperature variation is expected for $\tau_{th}$ which has very similar values in the low-temperature vortex solid (see below). We determine the liquid–solid transition temperature $T_m = 11.5\,\text{K}$ from the condition $\tau_{pl}(T_m)\omega_e = 1$:

$$T_m = \frac{T_c U_0}{U_0 - T_c \ln(\omega_e\tau_{th})}. \quad (5)$$

The barriers $U_{pl}(T_m) = 160\,\text{K}$ for plastic deformation at $T_m$ are roughly one order of magnitude larger than $T_m$, a situation similar to what has been reported for high-$T_c$ cuprates [2, 11]. Since $U_0 \propto a_0 \propto 1/\sqrt{B_0}$ (see also [2, 4]) we can estimate the position of the melting line $B(T_m)$ in the phase diagram from this equation (green line in figure 1).

The fact that the relaxation rate above $T_m$ is larger than that calculated with this single barrier $U_{pl}$ and stays close to the maximum value for fluctuations of full static field amplitude with characteristic time $\tau_c \omega_e = 1$ might indicate the presence of energy barriers of up to $400\,\text{K}$ at $T_m$. Such a distribution of deformation barriers could be induced by the random pinning potential, but the quantitative analysis is beyond the scope of our simple model for the spectral density of the fluctuations.

4.2. Thermal fluctuations in the solid

In the solid the fluctuations of the local field are due to the thermal position fluctuations $\langle u^2 \rangle$ around the equilibrium positions of the FLL. The variance $\sqrt{\langle u^2 \rangle}$ is small compared to the flux line spacing $a_0$, a conventional criterion for the melting temperature being $c_L(T_m) = \sqrt{\langle u^2 \rangle}/a_0 \approx 0.1-0.2$ [3, 4, 29]. $1/T_2$ falling below the maximum rate corresponding to full field fluctuations in figure 3 is an indication of this reduced amplitude. We approximate the amplitude
of the fluctuating field in the solid by the full amplitude in the field profile $b_i$ (equation (3)) times the displacement relative to the lattice constant:

$$n b_s(T) \approx b_i(T) \frac{\langle u^2 \rangle}{a_0} = b_i(T) c_L(T).$$

We use the approximation for the amplitude of the position fluctuations in the case of overlapping flux lines derived by Brandt [3]:

$$\langle u^2 \rangle = \langle u^2 \rangle_{\text{loc}} \left[ \frac{\kappa}{\pi} \sqrt{2b} + \sqrt{1 + \frac{\kappa^2 b}{2(1-b)}} \right],$$

(7)

$b = n \Phi_0/B_{c2} \approx B_0/B_{c2}$ is the mean internal field relative to the upper critical field $B_{c2}(T)$. Replacing $n \Phi_0$ by the applied field is a very good approximation, as seen in figure 2. The fluctuation amplitude in the local pinning limit at thermal energy $k_B T$ is [30]

$$\langle u^2 \rangle_{\text{loc}} = k_B T \sqrt{\frac{n}{4\pi \epsilon_{66} c_{44}}}.$$  

(8)

The homogeneous tilt modulus is approximated by $c_{44} \approx B_0^2/\mu_0$, where we again replaced $n \Phi_0$ by the applied field. The shear modulus $c_{66} = c_{44}(1-b)^2/(8\kappa^2)$ is for the present field small compared to the homogeneous tilt modulus. $c_{66}$ depends via $B_{c2}$ on temperature and approaches zero with increasing temperature at the critical line $B_{c2}(T)$. In principle, the shear modulus is expected to vanish already at the liquid–solid transition $T_m$, but this transition is outside the scope of the linear theory of elasticity. There are, however, indications that the liquid–solid transition is of first order [2, 6]. Equation (7) might, therefore, give a reasonable estimate of the fluctuation amplitude even at temperatures close to $T_m$.

Calculating the Lindemann number in equation (6) from these equations, we find $c_L(T_m) = 0.05$ and $c_L = 0.1$ is reached at $T = 12.4$ K. This is in reasonable agreement with the above-mentioned Lindemann criterion for the melting temperature since the fluctuation amplitude is usually underestimated by the theory of elasticity [3, 4]. The result is also consistent with the energy barrier $U_{\text{pl}}(T_m) = 160$ K for plastic deformation determined above. Comparing the exponential slowing down of the plastic mode with the Lindemann criterion, Blatter et al found [2] $T_m \approx 2c_{44}^2 U_{\text{pl}}(T_m)$, that is $c_L = 0.19$ for $T_m = 11.5$ K.

The temperature dependence of the shear modulus also determines that of the thermodynamic characteristic time in the vortex solid. The normal modes of the FLL are strongly overdamped, leading to exponential relaxation into its equilibrium positions [31, 32]. We focus on the relaxation of pure shear strain since this mode is much slower than the compressional mode:

$$\tau_{\text{th}} = \frac{\eta}{c_{66} k^2}.$$  

(9)

Here $\eta$ is the viscosity of the FLL per unit volume and $k$ is the lattice distortion wave vector (perpendicular to the flux lines for pure shear). For small wavelength distortions we substitute $k = \pi/a_0$. The viscosity can be approximated by $\eta = B_0^2/\rho_0 b$, where $\rho_0 = 0.13 \mu \Omega \text{m}$ is the electronic resistivity in the normal state [18] just above $T_c$. Within these approximations we find $\tau_{\text{th}} = 4.8 \times 10^{-10}$ s at $T = 11.5$ K. Considering the approximations involved, the value is very close to that estimated above for the liquid, another indication that extrapolations across
$T_m$ might work to some extent. The characteristic time of this diffusive mode decreases to low temperatures by a factor of 2 and it diverges at the critical line ($b = 1$).

Such very short time scales compared to the experimental ones are typical for the FLL and have been encountered in the discussion of $1/T_2$ in high-$T_c$ cuprates as well [11, 12]. With these characteristic times, small wavelength distortions are again completely ineffective for $1/T_2$. Equation (9) indicates, however, that for long-wavelength modes the characteristic time can reach $\tau_b = 1$. In our case of La$_3$Ni$_2$B$_2$N$_{3-x}$, this occurs at $k \approx 6.0 \times 10^{-4} \pi/a_0$, a value which below $T_m$ does not depend strongly on temperature. We ascribe the spin–spin relaxation at low temperatures to these modes and substitute in equation (2) $\tau_c = 1/\omega_c$. Inserting the fluctuating field amplitude from equation (6) and multiplying the Lindemann number by a constant $x_L$ to compensate for the low estimate of the fluctuations, we calculate the contribution of the solid FLL to the spin–spin relaxation (dotted blue in figure 3) with the single-fit parameter $x_L = 10.0$.

The very broad maximum in $1/T_2$ below $T_c/2$ calculated for these fluctuations is very similar to that calculated within the local pinning limit for YNi$_2$B$_2$C [8]. From the discussion above it should be clear, however, that the factor $x_L$ scaling the fluctuation amplitudes is restricted by the lattice constant of the FLL and cannot be used to fit $1/T_2$ in the upper temperature regime. In particular at the melting temperature, the Lindemann criterion implies that the fluctuation amplitude $(\gamma_b)\approx 0.04$ below the maximum value for fluctuations with the full amplitude. We therefore ascribe the large relaxation rate in the regime below $T_m$ to large fluctuation amplitudes in a crossover from long-wavelength shear modes to plastic deformation. We emphasize that this indicates an alternative origin for the two peaks in $1/T_2$ of YNi$_2$B$_2$C, which have been ascribed to a non-monotonic temperature dependence of the fluctuation amplitudes [8]. The narrow peak close to $T_c$ could be associated with plastic deformation, the low-temperature broad maximum with the shear mode. The absence of both in the intermediate temperature range might be explained by a loss of spectral weight of the shear mode at low frequency through shifting weight from extremely long to shorter wavelengths.

Finally, we briefly comment on the dependence of spin–spin relaxation on the thermal cycle during preparation of the samples. The discussion above strongly indicates that the sample with optimum microstructure is in the weak pinning regime—the random pinning potential was neglected throughout the discussion. Increasingly stronger pinning might explain the suppression of the flux line fluctuation contribution in the other two samples. Close to $T_c$ pinning will contribute to increased energy barriers for plastic deformation and shift this peak in $1/T_2$ closer to $T_c$, where the factor from the fluctuation amplitudes is quadratically smaller. At low temperatures stronger pinning could introduce a cut-off at several lattice distances for the shear mode, reducing the spectral density of the fluctuations at the frequency $\omega_c$ where the spin-echo mode at low frequency through shifting weight from extremely long to shorter wavelengths.

5. Conclusion

NMR spin-echo measurements of the inhomogeneous linewidth and the spin–spin relaxation rate of $^{11}$B in the field-cooled state of the type-II superconductor La$_3$Ni$_2$B$_2$N$_{3-x}$ agree for our cleanest sample and in the appropriate temperature ranges quantitatively with simple relaxation theory. Beyond the intrinsic Ginsburg–Landau penetration depth, coherence length, critical temperature $T_c(B = 0)$ and critical field $B_{c2}(T = 0)$, all of which are consistent with or taken from other independent measurements, the calculation contains only a single fit parameter to compensate for the low estimate of the position fluctuation amplitudes within elasticity theory.

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Our analysis of both, inhomogeneous linewidth and spin–spin relaxation, indicates that two characteristic time scales for large wavelength deformations should be taken into account. Upon cooling in a field $B_0 = 1.3$ T, the clean sample first enters a vortex liquid state at an onset $T_c = 12.7$ K. We find clear evidence in $1/T^2$ for the time scale associated with plastic deformation in a viscous liquid and determine from this the melting temperature $T_m = 11.5$ K as the temperature where the characteristic time of this process becomes longer than the time set by the spin-echo experiment. In the vortex solid at low temperature, we ascribe the fluctuating fields measured by $1/T^2$ quantitatively to long-wavelength shear modes of the vortex lattice. The extrapolation of the corresponding thermal vortex position fluctuations to large temperatures leads, in conjunction with the Lindemann criterion, to a melting temperature consistent with that determined from the liquid state. However, $1/T^2$ is underestimated significantly in the crossover regime below $T_m$, indicating that here a more complex model is required. The same two time scales may dominate the spin–spin relaxation from vortex fluctuations of other type-II superconductors in the weak pinning regime, and might be a natural explanation for the complex temperature dependence reported e.g. for $1/T^2$ in YNi$_2$B$_2$C crystals.

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