Four-fold basal plane anisotropy of the nonlocal magnetization of YNi$_2$B$_2$C

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Borocarbide superconductors have received considerable recent attention, due in part to the interaction between magnetism and superconductivity. A rich superconducting phase diagram, including transitions between hexagonal, rhombohedral, and square vortex lattices, has been observed. The existence of vortex lattices with non-hexagonal symmetry has been attributed to non-locality effects on the superconducting electrodynamics, which arise from the large electronic mean free path of these clean superconductors. Geometrically, a vortex directed along the tetragonal c-axis has current contours that are square-like. It has been shown in the non-magnetic borocarbide YNi$_2$B$_2$C, that the deviations from the standard (local) London magnetic field dependence of the equilibrium magnetization $M_{eq} \propto \ln(H)$ can be quantitatively accounted for by introducing non-local electrodynamics into the London description. Traditionally, it was widely thought that nonlocality effects should be significant only in materials with a Ginzburg-Landau parameter $\kappa \sim 1$; however, YNi$_2$B$_2$C has $\kappa \approx 10$ - 15.

In the local London model of superconducting vortices, the material anisotropy is introduced via a second rank mass tensor $m_{ij}$. In tetragonal materials such as YNi$_2$B$_2$C or LuNi$_2$B$_2$C, the masses in both principal directions in the square basal plane are the same, $m_a = m_b$, thus the superconducting properties are isotropic in the a-b plane. In contrast, non-local corrections are expected to introduce a four-fold anisotropy as a function of the magnetic field orientation within the a-b plane. A temperature dependent in-plane anisotropy of the upper critical field $H_{c2}$ has been observed in the non-magnetic borocarbide LuNi$_2$B$_2$C and described within a Ginzburg-Landau framework incorporating non-local effects. However, a direct observation of the in-plane anisotropy deep into the superconducting phase, where the non-local London model applies and unusual vortex lattices are observed, has not been reported until now.

In this work we show that, in the superconducting mixed state of YNi$_2$B$_2$C, the reversible magnetization oscillates with a $\pi/2$ periodicity when the applied field is rotated within the a-b plane. The amplitude of the angular oscillation decreases with field, passes through zero and then reverses sign at a field well below $H_{c2}$. The results are in good quantitative agreement with the non-local London description introduced by Kogan et al.

The 17 mg single crystal of YNi$_2$B$_2$C investigated in this study is the same as that previously used by Song et al. to explore the magnetic response when the applied field $H$ is parallel to the c-axis. The critical temperature is $T_c = 14.5$K, defined as the point at which the linearly varying magnetization $M(T)$ extrapolates to zero; this ignores a slight "tail" extending to 15.6K. The crystal is a slab of thickness $t \sim 0.5mm$, whose shape and size in the basal plane are sketched in Fig. 1. It will be useful to approximate such shape by an ellipse of axis $L_x$ and $L_y$. X-ray diffraction shows that the crystallographic c-axis is normal to the slab, and that the two equivalent (110) axes of the tetragonal structure very approximately coincide with the axes of the ellipse.

Measurements were performed in a Quantum Design SQUID magnetometer with a 50 kOe magnet. Two sets of detection coils allows us to measure both the longitudinal (parallel to $H$) and transverse (perpendicular to $H$) components of the magnetization vector $M$, but only the longitudinal component (denoted hereafter as simply the magnetization $M$) will be discussed in this work. The crystal was mounted into a previously described homemade rotating sample holder with rotation axis perpendicular to $H$. The c-axis was aligned with the rotation axis, so that $H$ could be rotated within the basal plane.

Figure 1 shows $M$ as a function of the angle $\varphi$ between $H$ and the a-axis (see sketch in Fig. 1), at $T = 7$K and two values of $H$. The crystal was initially cooled in zero field, $H$ was then applied and the sample was subsequently rotated in steps of $\Delta \varphi \approx 3.1^\circ$. Each data point was taken at fixed $\varphi$.

We first discuss the low field curve of fig. 1b. As $H = 300Oe$ is well below the lower critical field $H_{c1}$ for all $\varphi$, this curve represents the total flux exclusion of the Meissner state. The oscillatory behavior with periodicity $\pi$...
(two-fold symmetry) originates from geometrical effects. Indeed, a field applied at any orientation within the basal plane can be decomposed in \( H_x = H \cos(\varphi + 45^\circ) \) and \( H_y = H \sin(\varphi + 45^\circ) \). If we approximate the crystal shape by the ellipse, the Meissner response associated with each component is \( 4\pi M_i = -H_i/(1 - \nu_i) \), where \( i = x; y \) and \( \nu_i = t/L_i \) are the demagnetizing factors, thus \( 4\pi M = -H [\cos^2(\varphi + 45^\circ)/(1 - \nu_x) + \sin^2(\varphi + 45^\circ)/(1 - \nu_y)] \). The best fit to this expression gives \( \nu_x \sim 1/4 \) and \( \nu_y \sim 1/5 \). This corresponds to the ellipse of axes \( L_x \sim 2.0 \text{mm} \) and \( L_y \sim 2.5 \text{mm} \) shown in the sketch of fig. 1.

We now turn to the high field data of Fig. 1a. The applied field, \( H = 45kOe \), is well above \( H_{c2} \sim 35 \text{kOe} \) at this temperature (which is only weakly \( \varphi \) dependent, see below), thus in this case \( M(\varphi) = M^{eq}(\varphi) \) is the normal state paramagnetic response. We again observe an oscillatory behavior, but in this case the periodicity is \( \pi/2 \). By combining the information provided by the X-rays with the geometrical effects on the Meissner response, we conclude that the maximum normal state magnetization occurs at the crystallographic orientations (110) and \((\overline{1}00)\), while the minimum corresponds to (100) and \((010)\). No hint of the geometry-originated two-fold symmetry is observed. This is to be expected, as demagnetizing effects vanish in the limit \( |M/H| < 1 \). Further analysis of \( M^{ns} \) suggests that it arises from a low concentration, \( \sim 0.001 \) mol fraction, of rare earth ions, most likely from impurities in the yttrium starting metal. As shown below, \( M^{ns} \) is much smaller than the superconducting contribution except close to \( H_{c2} \).

The above procedure cannot be used in the superconducting mixed state, due to the appearance of magnetic hysteresis arising from vortex pinning. The critical current density \( J_c \) is very small in this crystal. As a result, the magnetic hysteresis \( (M^4 - M^7) \propto J_c \), where \( M^4 \) and \( M^7 \) are respectively the magnetizations measured in the field-decreasing and field-increasing branches of an isotermal \( M(H) \) loop, is small as compared to the equilibrium or reversible magnetization, \( M_{eq} \approx (M^4 + M^7)/2 \).

In spite of this, the residual hysteresis strongly affects the response obtained by rotating the crystal at fixed \( T \) and \( H \), by superimposing a periodicity \( \pi \) (related to shape effects on the critical state magnetization) that almost completely hides the intrinsic \( \pi/2 \) periodicity of fundamental interest.

To solve this difficulty, we performed magnetization loops at \( T = 7K \) at a set of fixed angles and then calculated \( M_{eq}(H) \) for each \( \varphi \). In all cases we extended the loops up to \( H = 50kOe \), thus we could repeat the measurement of \( M^{ns} \) in the normal state and compare the data with those obtained by rotating at fixed \( H \). Due to the absence of hysteresis, both determinations of \( M^{ns}(H, \varphi) \) should coincide. This is indeed the case as seen in Fig. 1a, where the open circles represent the data at \( H = 45kOe \) obtained from the \( M(H) \) loops.

Figure 2 shows \( M_{eq} \) (obtained from averaging \( M^4 \) and \( M^7 \)) as a function of \( \varphi \) for several \( H \). All the data have the same scale, but the curves at different \( H \) have been vertically shifted to accommodate the whole field range within the plot. For \( H < 1.5kOe \), the irreversibility becomes large enough to introduce a significant uncertainty in the determination of \( M_{eq} \), consequently those data have been disregarded. It is apparent that a four-fold symmetry exists in the whole field range of the measurements. To quantify the amplitude of the oscillations, we fitted the curves by \( M_{eq}(H, \varphi) = (M_{eq}) + \delta M_{eq}(H) \cos(4\varphi) \).

The values of \( \delta M_{eq}(H) \) so obtained are plotted in Figure 3, while the values of \( (M_{eq}) \) are shown in the inset. A remarkable fact, clearly visible in figs. 2 and 3, is that \( \delta M_{eq}(H) \) crosses zero and it reverses sign at some intermediate field (\( \sim 12 \text{kOe} \)) well within the superconducting mixed state. Another interesting observation is that the amplitude of the oscillations at \( H \sim 1.5 - 2 \text{kOe} \) is as large as that at \( H \sim 50kOe \).

The above results show that a \( \pi/2 \) basal plane anisotropy exists both in the normal and superconducting states. It is also clear from Fig. 3 that a change in the behavior of \( \delta M_{eq}(H) \) takes place at the superconducting transition at \( H_{c2} \sim 35kOe \). This observation, together with the sign reversal and the large amplitudes at low fields, point to the existence of a second source of in-plane anisotropy, in addition to the normal state one, that turns on in the superconducting phase.

Prior to analyzing the superconducting basal-plane anisotropy it is necessary to subtract the normal state contribution, which persists within the superconducting phase. To that end we performed rotations at fixed \( H \), as those shown in Fig. 1a, at several \( T \) and \( H \) above \( H_{c2}(T) \).

We found a paramagnetic response that exhibits a four-fold symmetry, with the minimum at \( \varphi = 0 \) in all cases, i.e., \( M^{ns}(\varphi = 45^\circ) > M^{ns}(\varphi = 0^\circ) > 0 \) for all \( T \) and \( H \). We thus have a well defined set of data \( \delta M^{ns}(H, T) \) which exhibits no sign reversal. The extrapolation is not obvious, however, as \( \delta M^{ns} \) is not linear in \( H \). Figure 4 shows all the \( \delta M^{ns} \) data collected at various temperatures \( 5K \leq T \leq 16K \) and \( H \geq 50kOe \), as a function of \( H/T \). We found that, when plotted in this way, all the data points collapse on a single curve.

The dashed line in Fig. 4 is a fit to the \( \delta M^{ns}(H/T) \) data. The same fit, for the case \( T = 7K \), is also shown as a dashed line in Fig. 3. We can now subtract that curve from the total \( M_{eq} \) shown in Fig. 3, to isolate the superconducting contribution \( \delta M_{sc} \).

Note that, as \( \delta M^{ns} \) is always positive and increases monotonically with \( H \), both the sign reversal and the non-monotonic behavior immediately below \( H_{c2} \) exhibited by \( M_{eq} \) must arise from the \( \delta M_{sc} \) contribution.

We now show that the four-fold symmetry of \( M_{eq}^{sc} \), as well as the field dependence of \( \delta M_{eq}^{sc} \), can be well described using the non-local modifications to the London electrodynamics introduced by Kogan et al. According to that model, for \( H_c \ll H \ll H_{c2} \),

\[
M_{eq}^{sc} = -M_0 \left[ \ln \left( \frac{H_0}{H} + 1 \right) - \frac{H_0}{H_0 + H} + \zeta \right] \quad (1)
\]
Here $M_0 = \Phi_0 / 32 \pi^2 \lambda^2$, the new characteristic field \( H_0 = \Phi_0 / 4 \pi^2 \rho^2 \) is related to the nonlocality radius \( \rho \), and \( \zeta = \eta_1 - \ln(H_0/\eta_2 H_{\text{eq}} + 1) \), where \( \eta_1 \) and \( \eta_2 \) are constants of order unity.

Song et al. have shown that the magnetization of this same crystal is very well described by Kogan’s model, when \( H \parallel c \)-axis. A fingerprint of the nonlocality effects is the deviation from the \( M_{\text{eq}} \propto \ln(H) \) behavior predicted by the local London model. The curvature clearly visible in the inset of Fig. 3 is thus a strong indication that nonlocality also plays a major role when \( H \perp c \)-axis. It is worth mentioning that, although a quantitative analysis of the curve in the inset of Fig. 3 in terms of Eq. 1 would require removal of the normal state magnetization, its contribution is small and would not significantly modify the curvature seen in the \( M_{\text{eq}} \) vs. \( \ln(H) \) data.

We now apply Eq. 1 to the analysis of our data. In principle, the basal plane anisotropy could be ascribed to the material parameters \( M_0, H_0 \) and \( H_{\text{eq}} \). However, \( M_0 \propto \lambda^{-2} \) is isotropic within the \( a-b \) plane of a tetragonal structure. On the other hand, four-fold variations of \( H_{\text{eq}} \) within the basal plane have been observed in LuNi$_2$B$_2$C and attributed to nonlocality. We then assume that both \( H_0 \) and \( H_{\text{eq}} \) have \( \pi/2 \) periodicity, \( H_0(\phi) = \langle H_0 \rangle + \delta H_0 \cos(4 \phi) \) and \( H_{\text{eq}}(\phi) = \langle H_{\text{eq}} \rangle + \delta H_{\text{eq}} \cos(4 \phi) \). To first order in \( \delta H_0 \) and \( \delta H_{\text{eq}} \) we obtain

\[
\delta M_{\text{eq}}^c = -M_0 \left[ \frac{1}{1 + \frac{H}{(H_0)}^2} - \alpha \right] \epsilon_1 + \alpha \epsilon_2
\]

where \( \alpha = \frac{1}{1 + \eta_2 \frac{H_0}{(H_0)}} \); \( \epsilon_1 = \frac{\delta H_0}{(H_0)} \); \( \epsilon_2 = \frac{\delta H_{\text{eq}}}{(H_{\text{eq}})} \).

Experimentally we have determined \( \langle H_{\text{eq}} \rangle = 35 \) kOe and \( \delta H_{\text{eq}} \sim 0.4 \) kOe (at \( T = 7K \)), so we can fix \( \epsilon_2 = 0.01 \). We could also attempt to determine \( M_0 \) and \( \langle H_0 \rangle \) by fitting our \( M_{\text{eq}} \) data with Eq. 1. However, this is a difficult task that requires the measurement of a large set of temperatures to check the consistency of the results. Instead, we decided to use the results of Song et al. (for \( H \parallel c \)-axis) as good estimates. For \( T = 7 \) K, we take \( \langle H_0 \rangle = 56 \) kOe and \( M_0 = 5.2 \) G. (Here we scaled down \( M_0 \propto 1/\lambda_0 \lambda_c \) by the experimental mass anisotropy, \( \gamma \approx 1.15 \), between the \( c \)-axis and the \( a-b \)-plane, which is close to the value \( \gamma \sim 1.1 \) obtained from band structure calculations.) In any case, small variations in any of these parameters will not significantly affect the rest of the analysis. If we also assume \( \eta_2 \sim 1 \), we obtain \( \alpha \sim 2/3 \). With these fixed parameters, we fit our \( \delta M_{\text{eq}}^c(H) \) data with Eq. 3 with the single free parameter \( \epsilon_1 \). We obtain \( \epsilon_1 = 0.14 \). The fitted curve (for \( \delta M_{\text{eq}}^c + \delta M_{\text{mag}}^c \)) is shown as a solid line in Fig. 3.

The very good coincidence between our data and the model is remarkable. With a single fitting parameter \( \epsilon_1 \), which is field independent, we have been able to account for the nontrivial \( H \) dependence of \( \delta M_{\text{eq}}^c \), including the sign reversal at intermediate fields. Of course, the fit deviates from the data close to \( H_{\text{eq}} \), where the London model fails. These experimental results show that nonlocality effects have a profound effect on these clean, intermediate-\( \kappa \) superconductors and they underscore the remarkable utility of the generalized London theory.

In summary, we have demonstrated a four-fold anisotropy in the square basal plane of clean single crystal YNi$_2$B$_2$C. This superconducting response is inconsistent with conventional local London theory, but it is well explained by a generalized London model incorporating non-local electrodynamics, with parameters based largely on complementary experiments. These observed effects of nonlocality persist deep into the superconducting state, where complex, evolving vortex lattices occur.

[Note added: while preparing this manuscript, we learned that P. C. Canfield et al. at the Ames Lab. have observed similar oscillations of the basal plane magnetization of LuNi$_2$B$_2$C.]

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FIG. 1. Equilibrium magnetization $M_{eq}$ at $T = 7K$, as a function of the angle $\phi$ between the applied field $H$ (contained in the $a$-$b$ plane) and the $a$-axis, for (a) $H = 45kOe$ and (b) $H = 300Oe$. The inset shows a sketch of the crystal shape and crystallographic orientation in the basal plane, superimposed to an ellipse with the same demagnetizing factors.

FIG. 2. Reversible magnetization $M_{eq}$ at $T = 7K$, as a function of the angle $\phi$ between the applied field $H$ (contained in the $a$-$b$ plane) and the $a$-axis. The fields (in $kOe$) are indicated next to each curve. The scale is the same for all the curves, but data at different $H$ have been vertically shifted.

FIG. 3. Amplitude $\delta M_{eq}$ of the four-fold oscillations of the basal plane magnetization, as a function of the applied field $H$. The dashed line is the normal state contribution $\delta M^{ns}$ obtained from the fit shown in Fig. 4. The solid line is the fit to $\delta M^{eq} + \delta M^{ns}$ using Eq. (2) with $\epsilon_1$ as the only fitting parameter. Inset: average in-plane magnetization $\langle M_{eq} \rangle$ as a function of applied field (see text).

FIG. 4. Amplitude $\delta M^{ns}$ of the four-fold oscillations of the basal plane magnetization in the normal state, as a function of $H/T$. The dashed line is a polynomial fit.
