In cuprate (high-$T_c$) superconductors, high-transition temperatures ($T_c$) and short coherence lengths ($\xi$) lead to large thermal fluctuation effects, opening a possibility for melting of the flux line lattice (FLL) at temperatures well below the superconducting transition temperature. A discontinuous step in $dc$ magnetization and a sudden, kink-like drop in resistivity signified the first order nature of the melting transition from the vortex lattice into a liquid. In conventional type II superconductors, with modest transition temperatures and large coherence lengths, vortex melting is also expected to occur in a very limited part of the phase diagram but it has yet to be observed experimentally. In the rare-earth nickel borocarbides RNi$_2$B$_2$C ($R = $ Y, Dy, Ho, Er, Tm, Lu), the coherence lengths ($\xi \approx 10^2 A$) and superconducting transition temperatures ($16.1\,K$ for $R = $ Lu) lie between these extremes, suggesting that the vortex melting will be observable and may provide further information on vortex dynamics. Indeed, Mun et al.\textsuperscript{12} reported the observation of vortex melting in YNi$_2$B$_2$C, based on a sharp, kink-like drop in electrical resistivity.

Recently, a magnetic field-driven FLL transition has been observed in the tetragonal borocarbides\textsuperscript{6,7,8,9} The transition from square to hexagonal vortex lattice occurs due to the competition between sources of anisotropy and vortex-vortex interactions. The repulsive nature of the vortex interaction favors the hexagonal Abrikosov lattice, whose vortex spacing is larger than that of a square lattice. The competing anisotropy, which favors a square lattice, can be due to lattice effects (fourfold Fermi surface anisotropy)\textsuperscript{10} unconventional superconducting order parameter\textsuperscript{11} or an interplay of the two.\textsuperscript{12,13} In combination with non-negligible fluctuation effects, the competition leads to unique vortex dynamics right below the $H_{c2}$ line in the borocarbides, namely a reentrant vortex lattice transition. Fluctuation effects near the upper critical field line wash out the anisotropy effect, stabilizing the Abrikosov hexagonal lattice.\textsuperscript{14,15} Here, we report the first observation of paramagnetic effects in the $dc$ magnetization $M$ of the mixed state of LuNi$_2$B$_2$C. The kink-like feature in $M$ and the corresponding specific heat feature for $H \geq 30\,kOe$ signify the reentrant FLL transition, which is consistent with the low-field FLL transition line inferred from small angle neutron scattering (SANS).\textsuperscript{16}

Single crystals of LuNi$_2$B$_2$C were grown in a Ni$_2$B flux as described elsewhere and were post-growth annealed at $T = 1000\,C$ for 100 hours under high vacuum, typically low $10^{-6}\,Torr$. Samples subjected to a preparation process such as grinding, were annealed again at the same condition as the post-growth annealing. A Quantum Design magnetic property measurement system (MPMS) was used to measure $ac$ and $dc$ magnetization while the heat capacity option of a Quantum Design physical property measurement system (PPMS) was used for specific heat measurements. Electrical resistivity was measured by using a Linear Research $ac$ resistance bridge (LR-700) in combination with a PPMS. The detailed $dc$ magnetization of LuNi$_2$B$_2$C reveals an anomalous paramagnetic effect for $H \geq 30\,kOe$, where the magnetic response deviates from a monotonic decrease and starts to rise, showing decreased diamagnetic response. The in-phase and out-of-phase components of the $ac$ susceptibility $\chi_{ac}$ show a dip and the specific heat data show a feature at the corresponding temperature, reminiscent of vortex melting in high-$T_c$ cuprates.\textsuperscript{17} Electrical transport measurements, however, do not exhibit any feature corresponding to the paramagnetic effect; $e.g.$, a sharp drop in the electrical resistivity. The zero-resistance transition, rather, occurs at a much higher temperature, suggesting that the paramag-
netic effect is not related to vortex melting. It is instead consistent with a topological FLL change between square and hexagonal structures.

The top panel of Fig. 1 shows dc magnetization $M$ as a function of temperature at several fields. For $H \geq 30$ kOe, kink-like features appear, which are marked by arrows. The anomalous increase can be easily seen as a sharp drop in $dM/dT$ (arrows in the bottom panel). The magnetization reported here is independent of time and has no hysteresis between zero-field cool (ZFC) and field cool (FC) data within experimental accuracy, indicating that the measured value is an equilibrium magnetization. In the top panel of Fig. 1, we compare the data and some model calculations. Dashed lines are predictions from the standard local London model\cite{note} and solid lines from the non-local London model\cite{note} (see text). Bottom panel: temperature derivation of $M(T)$ at corresponding magnetic fields. Arrows indicate the points where kink-like features start to appear.

\[ -4\pi M = M_0 \ln\left(\frac{\eta H_{c2}}{H}\right), \quad (1) \]

where $M_0 = \phi_0 / 8\pi\lambda^2$, $\eta$ is a constant of order unity, $\lambda$ the penetration depth, and $\phi_0$ the flux quantum. In the fit, $H_{c2}$ was determined from our resistivity data (see Fig. 2) and $M_0$ from $H_{c2}$ with $\kappa = \lambda / \xi = 15$. In order to get the best result, the fitting parameter $\eta$ was varied between 0.95 and 0.97 and the absolute amplitude of $M_0$ was changed as a function of magnetic field. The local London model explains the monotonic decrease with decreasing temperature, but the fit becomes worse at higher field. In a clean system like LuNi$_2$B$_2$C where the electronic mean free path is long compared to the coherence length $\xi_0$, the current at a point depends on magnetic fields within a characteristic length $\rho$, or nonlocal radius. Taking into account the nonlocal current-field relation in superconductors, a non-local London model was suggested:\cite{note}

\[ -4\pi M = M_0[\ln(1 + H_0/H) - H_0/(H_0 + H) + \Lambda], \quad (2) \]

where $H_0 = \phi_0 / (4\pi^2\rho^2)$ and $\Lambda = \eta_1 - \ln(1 + H_0/\eta_2 H_{c2})$ with $\eta_1$ and $\eta_2$ being order of unity. It is worth noting that the scaling parameter $H_{c2}$ in the local theory is replaced by $H_0$ in the non-local model. The nonlocal radius $\rho$ slowly decreases with increasing temperature and is suppressed strongly by scattering. The solid lines are best results from the model calculation where we used the temperature dependence of $H_0$ and $\Lambda$ from the literature for YNi$_2$B$_2$C.\cite{note} Both the local and the non-local models explain the temperature dependence of $M$ at low fields, while only the non-local model can describe the data at and above 35 kOe. The good fit from the nonlocal model is consistent with the equilibrium magnetization analysis of YNi$_2$B$_2$C.\cite{note} suggesting the importance of nonlocal effects in the magnetization. The many fitting parameters in both fits, however, prevent us from making a definite conclusion as to which model better describes $M(T)$. Nevertheless, we can extract the important conclusion that the kink-like feature in the mixed state is a new phenomenon that needs further explanation.

In the early stage of high-$T_c$ cuprate research, anomalous paramagnetic effects in $M(T)$ were reported in the irreversible region and this effect was later attributed to the field inhomogeneity of the measured scan length in a SQUID magnetometer.\cite{note} We tested various scan lengths from 1.8 cm to 6 cm for which the field inhomogeneity varies from 0.005 % to 1.4 % along the scan length and found negligible dependence on the measuring length, which suggests that field inhomogeneity is not the source of the anomaly. A more definitive test used a conventional type II superconductor NbSe$_2$ in a similar configuration. There was no such anomalies in NbSe$_2$ as in the borocarbide. Taken together, we conclude that the reversible paramagnetic effects are intrinsic to LuNi$_2$B$_2$C. We also emphasize that the the phenomena is different from the paramagnetic Meissner effect (PME) or Wohlfleben effect where the FC $\chi$ becomes positive whereas the ZFC $\chi$ remains negative. The PME is an irreversible effect and occurs in the Meissner state, while the subject of this study is a reversible effect and takes place in the mixed state.

In the top panel of Fig. 2 the reversible magnetization $M$ (left axis) and the out-of-phase component of ac susceptibility $\chi''$ (right axis) are shown as a function of
temperature at 40 kOe. A dip appears both in χ′ ac (not shown) and in χ″ ac at the same temperature where M shows the paramagnetic anomaly. Since a dip in χ ac is often related to vortex melting, it is natural to consider the vortex phase change from liquid to lattice or glass as a possible explanation. The resistive superconducting transition at 40 kOe (circles) and 50 kOe (squares) is shown in the bottom panel of Fig. 2. A resistive slope change in the transition region, that can be considered as a signature of the vortex melting line, was observed at 8.1 K and 6.6 K for 40 kOe and 50 kOe, respectively. The R = 0 transition temperature, however, is much higher than the temperature where the dip occurs in χ ac, which argues against the vortex melting scenario as the physical origin of the anomalous paramagnetic effects. The increase in M at the transition temperature is also opposite from the decrease in the vortex melting interpretation.

Recently, a structural phase transition in the FLL was suggested to explain another peak effect observed below the vortex melting line in YBCO.24,25 The vanishing of a squash elastic mode gives rise to a topological FLL transition and leads to the new peak effect, while the softening of the shear modes c66 is relevant to the conventional peak effect in high-Tc cuprates.26 The observation of the dip effect well below the melting line in LuNi2B2C indicates that the anomalous paramagnetic effects are related to a change in the FLL and the increase in M is also consistent with the FLL change where the Abrikosov geometrical factor β changes.20,27 Fig. 3 shows the temperature dependence of the paramagnetic anomaly, ∆(4πM), at several magnetic fields, where ∆(4πM) is the magnetization after subtracting the monotonic, diamagnetic background obtained from Eq. (2). With increasing field, the peak becomes enhanced and an additional peak is observed at 45 kOe. In extreme type II material (κ >> 1/√2), the magnetization change due to a FLL transition is written as

$$\Delta(4\pi M) = \frac{H_{c2} - H}{2\kappa^2 - 1} \left( \frac{1}{\beta_\Delta} - \frac{1}{\beta_0} \right),$$

where βΔ ≈ 1.16 for a hexagonal FLL and βΔ ≈ 1.18 for a square FLL. In the inset of Fig. 3, we compared the peak intensity of ∆(4πM) (left axis) and the estimation from Eq. 3 (right axis) with κ = 15. It is encouraging to see that the simple model qualitatively reproduces the field dependence of the paramagnetic contribution. However, the quantitative difference in absolute values suggests that a more elaborate model is required.

The H − T phase diagram is shown in Fig. 4. The upper critical field line Hc2 was determined from the R = 0 superconducting transition and is consistent with the temperature where χ ac starts to have a non-zero value. The H1 line in the mixed state is the point where the reversible magnetization shows the paramagnetic effects and H2 is the 2nd anomaly that appears above 45 kOe (see Fig. 1). According to the Gurevich-Kogan non-local London model, the anisotropic nonlocal potential, which is responsible for the low-field FLL transition observed in SANS, is averaged out by thermal vortex fluctuations near Hc2. Since the interaction becomes isotropic, the hexagonal Abrikosov lattice is preferable, leading to the second FLL transition from square back...
the line. The dotted line is the FLL transition line that meets the rhombic (triangular) lattice as the field gets closer to square and rhombic shapes are forms of vortex lattices. The prediction is consistent with our observation that the nonlocal effects is much smaller than that for the vortex melting. This prediction is consistent with our observation that the nonlocal effects is much smaller than that for the vortex melting. The dashed line depicts qualitatively what fluctuation effects. We note that a direct comparison between the SANS and our data is difficult even in resolving the issue. Finally, we note that we are not able to discern any corresponding feature to the H2 line in C_p or in χ_{ac}. More work is in progress to understand the second paramagnetic jump in M which appears for H ≥ 45 kOe.

In summary, we report the first observation of an anomalous paramagnetic jump in the magnetization of the mixed state of LuNi2B2C. A dip appears in χ_{ac} at the same temperature as the paramagnetic effects, suggesting the relevance of the flux line lattice. The H − T phase diagram is consistent with a FLL structural transition from square to hexagonal lattice just below the upper critical field line. The observation of an additional feature in the specific heat data at the corresponding temperature underscores the interpretation of paramagnetic effects as due to a reentrant FLL transition in LuNi2B2C.

Work at Los Alamos was performed under the auspices of the U.S. Department of Energy (DOE) and at Urbana under NSF Grant No. DMR 99-72087. The work at Pohang was supported by the Ministry of Science and
Technology of Korea through the Creative Research Initiative Program and at Ames by Iowa State University of Science and Technology under DOE Contract No. W-7405-ENG-82. We acknowledge benefits from discussion with Lev N. Bulaevskii, M. P. Maley, and I. Vekhter. We thank T. Darling for assistance in sample annealing.

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