Magnetic resonance imaging of laser polarized liquid xenon


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We demonstrate magnetic resonance imaging (MRI) of laser polarized liquid xenon, and image exchange between the liquid and vapor phases. The exceptionally large magnetization density of this liquid should allow MRI with micron-scale spatial resolution without signal averaging. Applications may include imaging of density equilibration and convective flow near xenon’s liquid-vapor critical point, low-field imaging of porous media microstructure, and mapping of the dynamics of two-phase (liquid-gas) flows.

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Xenon is unique among the noble gases, remaining liquid near room temperature: its liquid-vapor critical point is at \( \sim 290 \) \( K \) and 58 atm, and its solid-liquid-vapor triple point is at \( \sim 161 \) \( K \) and 0.8 atm. Liquid xenon has been employed in a variety of fundamental scientific investigations. For example, light scattering experiments near xenon’s critical point have served as powerful probes of universal phenomena such as the range and lifetime of order parameter fluctuations [1], as well as the “critical slowing down” of diffusive and dissipative processes [2].

Xenon is nearly inert chemically, but because of the xenon atom’s large electric polarizability the liquid is an excellent solvent—particularly for aliphatic materials. For example, hexadecane, benzene, toluene, hexane, and dodecane all have solubilities \( > 10\% \) in liquid xenon at \( \sim 0 \) \( ^{\circ} C \) [3]. In addition, nuclear magnetic resonance (NMR) studies of the spin-\( \frac{3}{2} \) 129Xe isotope have measured a relatively constant spin polarization lifetime \( (T_1) \) of \( \sim 25 \) min [4,5] and a relatively long spin decoherence time \( (T_2) > 1 \) s throughout the liquid xenon phase diagram [6], to within a few millikelvin of the critical point [7,8], and into the supercritical regime [9]. Thus liquid xenon approaches the behavior of an ideal, inert, NMR-detectable solvent.

Recently, Sauer, Fitzgerald, and Happer created laser polarized liquid xenon [5] by condensation of laser polarized xenon gas (i.e., gas with the 129Xe component nuclear spin polarized via spin-exchange optical pumping [10]). Laser polarized liquid xenon can have a very large magnetization density, approximately \( 10^3 \) times greater than that of thermally polarized liquids such as water in magnetic fields of a few tesla. This exceptionally large magnetization density enables a variety of interesting scientific and technical applications, including: enhancement of the nuclear spin polarization of molecules dissolved in liquid xenon [11] (e.g., to aid NMR molecular spectroscopy and quantum computing); efficient transport and storage of polarized xenon for use in gas phase magnetic resonance imaging (MRI) [5]; and, as reported in this paper, high resolution MRI of liquid xenon and imaging of chemical exchange between the xenon liquid and vapor phases. With further development, laser polarized liquid xenon MRI may allow imaging of density equilibration and convective flow near xenon’s liquid-vapor critical point, low-field imaging of porous media microstructure, and mapping of the dynamics of two-phase (liquid-gas) flows.

(Note: NMR techniques have been used previously to study laser polarized liquid 3He and its vapor [12] and liquid 3He-4He mixtures [13], at cryogenic temperatures \( \sim 1 \) \( K \). This work shows that one can distinguish between the NMR response of liquid and vapor phases, as well as obtain useful information about the geometry of the liquid phase and interphase dynamics [14]. In addition, three-dimensional gradient NMR techniques have been used to investigate the flow of liquid 3He-4He mixtures in complex geometries [15].)

As a demonstration, we performed MRI on a drop of \( \sim 50 \) mm\(^3\) of laser polarized liquid xenon residing in the corner of a closed cylindrical Pyrex cell of \( \sim 25 \) cm\(^3\) volume [see Fig. 1(a)]. A complimentary image of the laser polarized xenon vapor filling the remainder of the Pyrex cell is shown in Fig. 1(b). Laser polarized liquid xenon was created by condensation of 80 atm·cm\(^3\) of xenon gas with an enriched abundance (90%) of the 129Xe isotope, and a 129Xe nuclear spin polarization of \( \sim 5\% \) induced by standard spin-exchange optical pumping techniques [10]. The Pyrex cell was held in an iso-octane slush bath at 166 K [16], near the liquid xenon boiling point with an associated xenon vapor pressure of \( \sim 1 \) atm. The evaporation lifetime for the liquid xenon drop was comparable to the spin polarization lifetime (\( \sim 25 \) min) and much longer than the time needed to acquire a magnetic resonance image (\( \sim 10 \) s).

The images shown in Fig. 1 were obtained using a 4.7 T GE Omega/CSI spectrometer/imager operating at 55.3 MHz for 129Xe using a homebuilt solenoid RF coil. Standard non-slice-selective fast gradient echo techniques were used for acquiring the laser polarized xenon images. The magnetic field gradient coils of the Omega instrument provide a maximum gradient strength of only 7 G/cm, which significantly limits the achievable MRI spatial resolution, even for laser polarized liquid xenon with its very large magnetization density [17]. Thus using the Omega’s maximum gradient strength, we obtained a two-dimensional spatial resolution of 195×195 \( \mu \)m for the liquid xenon image in Fig. 1(a). Diffusive atomic exchange between the xenon vapor and liquid can also be inferred from the image in Fig. 1(b). This image was acquired with RF pulses exciting only the vapor reso-
The fast, ultrahigh resolution of laser polarized liquid xenon MRI may enable a variety of scientific and technical investigations. We discuss three examples here. First, this new imaging technique could be used to study the universal phenomenon of density equilibration in critical fluids. Recent theoretical work [21] indicates that when a near-critical fluid is exposed to an external perturbation such as a change in temperature of one end of the sample, an adiabatic equilibration mechanism known as the ‘‘piston effect’’ [22] induces transient density boundary layers in the fluid density. For xenon within about one degree of its liquid–vapor critical point these density boundary layers are predicted to be ∼50 μm thick, to deviate by ∼2% from the nominal density profile (a smooth gradient induced by gravity), and to persist for a few seconds [21]. Under certain nonequilibrium conditions, anomalously high density in the transient boundary layer may also induce convective flow in the critical fluid and significantly change the system’s dynamic behavior and transport properties. Laser polarized xenon MRI may be used to measure the density, thickness, and lifetime of such transient density boundary layers, and to image convective near-critical flow.
A second promising application of laser polarized xenon MRI is to investigate two-phase (liquid-gas) phenomena. As a demonstration, we imaged xenon spins evaporating from the liquid into the vapor (Fig. 2), as well as condensing from the vapor into the liquid (Fig. 3). These images were acquired using appropriate combinations of RF and magnetic field gradient pulses to selectively destroy the laser-polarization-produced magnetization in one phase (liquid or vapor), while leaving undisturbed the magnetization of the other phase. Sequential images were then acquired of the phase whose magnetization had been destroyed, showing the buildup of magnetization as polarized xenon atoms move between phases to re-establish equilibrium. The images in Figs. 2 and 3 represent the net transport of xenon magnetization due to three competing processes: (i) the slow boiling of the liquid xenon drop (lifetime ~25 min); (ii) intraphase and interphase atomic diffusion; and (iii) depletion of the xenon magnetization by the acquisition of images. Extending these MRI techniques to flowing two-phase systems may allow the simultaneous mapping of dynamics in both phases. For example, the polarized-liquid-induced distortion of the xenon vapor image shown in Fig. 1(b) indicates an effective coupling between the liquid and vapor phases, which may enable measurement of the relative velocity field of a two-phase flow using multidimensional NMR correlation techniques [23].

Finally, we note a third promising application of laser polarized liquid xenon: serving as a probe of porous media microstructure using low-field MRI. Conventional, high-magnetic-field MRI of thermally polarized liquids infused in porous media (reservoir rocks, ceramics, etc.) suffers from large, local magnetic field gradients caused by the magnetic susceptibility difference between the solid grains and the infused liquid. These field gradients can be as high as 1000 G/cm at typical magnetic fields of a few tesla [24], and thus can severely degrade and distort images of the sample pore space. Operating at lower magnetic fields (<0.1 T) greatly reduces the effect of magnetic susceptibility gradients (which scale as the applied field squared); however, very low spin polarization makes micron-scale MRI of thermally polarized samples impractical at low magnetic fields. In contrast, the magnetization of a laser polarized sample is independent of the applied field strength, which enables practical low-field imaging. We recently demonstrated MRI of laser polarized 3He gas at 21 gauss (with ~1 mm spatial resolution, limited by rapid gas diffusion), and showed the greatly reduced effects of magnetic susceptibility gradients [25]. Low-field MRI of infused laser polarized liquid xenon may provide direct visualization of porous microstructure on the micron scale, which at present is best inferred indirectly with NMR using pulsed-field-gradient diffusion measurements [26, 27].
In conclusion, we have demonstrated magnetic resonance imaging (MRI) of laser polarized liquid xenon, and imaged chemical exchange between the liquid and vapor phases. The exceptionally large magnetization density of this liquid should allow MRI with micron-scale spatial resolution without signal averaging. Applications may include imaging of density equilibration and convective flow near the liquid-vapor critical point, low-field imaging of porous media microstructure, and mapping of the dynamics of two-phase (liquid-gas) flows.

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