

Subfemtotesla radio-frequency atomic magnetometer for detection of nuclear quadrupole resonance

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A radio-frequency tunable atomic magnetometer is developed for detection of nuclear quadrupole resonance (NQR) from room temperature solids. It has a field sensitivity $0.24 \text{ fT/Hz}^{1/2}$ at the 423 kHz ^{14}N NQR frequency of ammonium nitrate. A potential application of the magnetometer is detection of nitrogen-containing explosives which is difficult with conventional tuned copper coils due to a poor signal-to-noise ratio (SNR) below a few megahertz. The NQR signal from 22 g of powdered ammonium nitrate located 2 cm away from the sensor is detected with a SNR of 9 in a 4.4-s-long multiple echo sequence, which represents an estimated order-of-magnitude improvement in sensitivity over the pickup coil detection. © 2006 American Institute of Physics.

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Optically pumped alkali metal vapor can be used as a resonant receiver of a radio-frequency (rf) magnetic field, with a sensitivity potentially surpassing that of conventional induction coils, especially at low frequencies.¹ The resonance is achieved by applying a static magnetic field B_0 which tunes the ground state Zeeman splitting of the alkali atoms to the signal frequency. In this arrangement, an oscillating rf signal of amplitude B_s applied perpendicular to B_0 induces a transverse polarization of the atomic spins given by $P_x \equiv F_x/F_z = (1/2)\gamma B_s T_2$ in the rotating frame. Here F is the total angular momentum of the atoms, γ is the gyromagnetic ratio, and T_2 is the transverse relaxation time of the polarization. The spin polarization oscillating in the laboratory frame is then measured by detecting the optical rotation of a linearly polarized laser beam passing through the vapor. Optical pumping of the atoms into the single Zeeman sublevel with the largest angular momentum projected along the pumping direction partially suppresses resonance broadening due to spin-exchange collisions.² This lengthening of T_2 is critical to achieve the high sensitivity of the magnetometer. Savukov *et al.*¹ demonstrated a potassium rf magnetometer with a sensitivity of $2 \text{ fT/Hz}^{1/2}$, limited by laser and ambient field noise. The calculated fundamental sensitivity, determined by quantum fluctuations, was at the level of $10^{-2} \text{ fT/Hz}^{1/2}$ for a cell with a volume of 200 cm^3 .

In this letter, we demonstrate a potassium rf magnetometer with a sensitivity of $0.24 \text{ fT/Hz}^{1/2}$ operating at 423 kHz. The magnetometer was used to detect a ^{14}N nuclear quadrupole resonance (NQR) signal from room temperature ammonium nitrate (NH_4NO_3) in the zero-applied field limit. Atomic magnetometers have recently been used to detect nuclear magnetic resonance from thermally polarized

water³⁻⁵ and laser polarized xenon^{3,6} with high sensitivity. Detection of NQR with no polarization enhancement is challenging because of the small size and short duration of the rf-induced signal. In light of potential applications of solid-state zero-field NQR in contraband detection, proposals have been made to improve upon the sensitivity of conventional pickup coil detection by using superconducting resonators⁷ and sensors (superconducting quantum interference devices)^{8,9} operating at cryogenic temperatures. Our results demonstrate the first detection of NQR with an atomic magnetometer and show that a cryogen-free atomic magnetometer, with intrinsically frequency-independent sensitivity and easy tuning/damping capabilities, could make an attractive new tool for detecting magnetic resonance signals in the kilohertz to megahertz range.

Figure 1 shows the experimental setup. A borosilicate rectangular glass cell, with an inner dimension of $40 \times 40 \times 60 \text{ mm}^3$, was filled with a drop of potassium metal, 70 torr of N_2 , and 630 torr of ^4He at room temperature. The cell was placed in a G-7 fiberglass oven which was heated to 180°C by hot air. The NQR sample was put in a high thermal-conductivity ceramic (boron nitride) container and was located on top of the oven, separated by a 6-mm-thick microporous insulation material. The separation between the bottom of the sample and the top of the K vapor was 2 cm. A continuous flow of chilled water in a plastic tube wrapped around the upper part of the boron nitride container kept the sample temperature at 24°C within $\pm 0.5^\circ\text{C}$. The sample and the oven were placed inside a single-layer aluminum and a double-layer mu metal shield; each layer of the shields had four holes in the xz plane for laser beam access [Fig. 1(b)]. Four evacuated, rectangular glass tubes were placed in contact with the K cell in order to secure turbulence-free paths for the laser beams.

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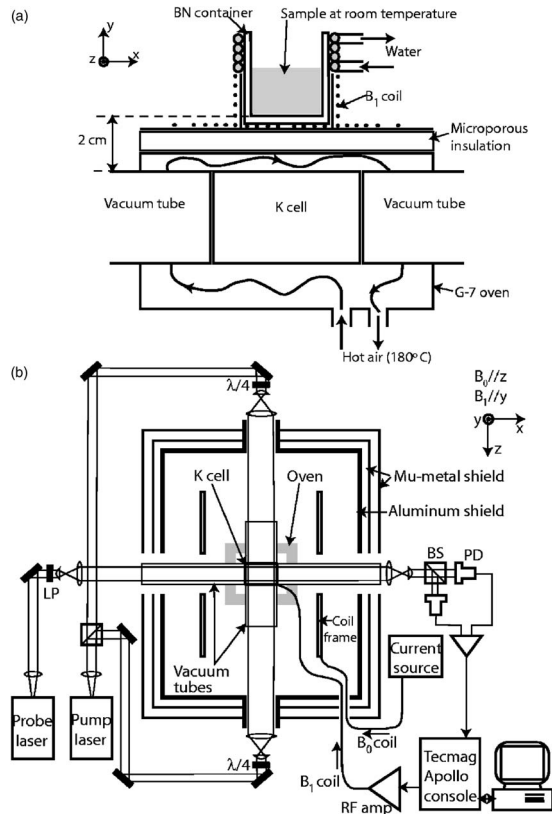


FIG. 1. (a) Schematic of the sample, boron nitride (BN) container, and the K cell inside a hot air oven. (b) Radio-frequency atomic magnetometer setup showing the pump and the probe beams, a rf (aluminum) and a static (mu metal) magnetic field shield, and balanced polarimeter. The offset field (B_{off}) coils, not shown, are directly wound on the two vacuum tubes along the pump beam direction close to the K cell. LP, linear polarizer; $\lambda/4$, quarter-wave plate; BS, beam splitter; PD, photodiode.

The K atoms were optically pumped to the $|F=2; m_F=2\rangle$ Zeeman sublevel under a static magnetic field $B_0 = B_0 \hat{z}$, by a high power (up to 400 mW) diode laser (Sacher Lasertechnik) tuned to the $D1$ line of potassium at $\lambda = 770$ nm. The cell was illuminated from both directions along the z axis. This arrangement of the pump beam is expected to help maintain a nearly uniform light intensity across an optically thick vapor cell. The transverse polarization P_x of the potassium atoms was measured by detecting the optical rotation of linearly polarized probe light using a balanced polarimeter. The probe beam was produced by a distributed feedback diode laser with a tapered amplifier (Toptica Photonics) at a wavelength $\delta\lambda \approx 0.1$ nm away from optical resonance. Typical probe beam power reaching the K cell was about 40 mW. Both the pump beam and the probe beam were expanded by factors of up to 40 by two-stage refractive optics involving planoconvex and aspheric lenses, which relied on spherical aberration to convert the initial Gaussian profile of the laser beam into a more uniform one over the illuminated volume of the cell.

Figure 2 shows the noise spectrum of the magnetometer which was tuned to $\omega_0/2\pi = 423$ kHz by $B_0 = \omega_0/\gamma_K = 0.604$ G. Here $\gamma_K = 2\pi \times 7.00 \times 10^5$ Hz/G is the gyromagnetic ratio of ground state potassium. The calibration field, which appears as a peak at the center, was applied uniformly over the cell. The noise level of 0.24 fT/Hz^{1/2} represents an eightfold improvement over the result in Ref. 1. The noise near resonance decreases by not more than 20% when the pump beam is shut off, which indicates that neither the pump

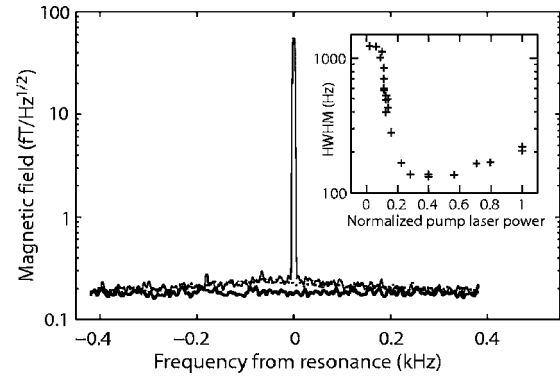


FIG. 2. Noise spectrum of the magnetometer obtained with (light solid line) and without (dotted line) a rf calibration field. The heavy solid line shows the noise when the pump beam was shut off. The photon shot noise corresponded to 0.1 fT/Hz^{1/2}. The inset shows the resonance half-width as the pump beam power was varied.

beam nor the external rf field fluctuation is the dominant source of the magnetometer noise. Instead, the frequency and intensity noise of the probe laser apparently are responsible for the detected noise, approximately two times larger than the shot noise. The inset of Fig. 2 shows the half-width at half maximum $\Gamma/2\pi = 1/(2\pi T_2)$ of the potassium Zeeman resonance as a function of the pump beam power. The resonance was measured by sweeping the applied rf field frequency at a constant B_0 . Resonance line narrowing by increased optical pumping is apparent in the low pump power regime. The linewidth eventually grows as optical pumping destroys transverse polarization of K generated by the rf field. In our setup the best magnetometer sensitivity was obtained with pump and probe beam intensities that produced $\Gamma/2\pi \sim 220$ Hz, which corresponded to a magnetometer Q value of 1000.

Free spin precession in NQR can be induced by a rf pulse applied in the same direction as the detection direction.¹⁰ For a resonant rf pulse applied to a spin-1 nucleus in a powdered sample, the induced signal varies with the “tipping angle” $\theta = \gamma_N B_1 t$ as $S \propto J_{3/2}(\theta)/\theta^{1/2}$.¹¹ Here B_1 and t are the rf pulse amplitude and length, respectively, and $J_{3/2}$ is the Bessel function of order 3/2. Here and in what follows we use subscript N to distinguish the gyromagnetic ratio and relaxation times of nuclear spins from those of K atomic spin. Since the gyromagnetic ratio of K is more than 2000 times larger than that of ¹⁴N, even a small fraction of the rf pulse amplitude that couples to the tuned magnetometer will easily saturate the magnetometer, diminishing the K polarization. To prevent this we have adopted two techniques. First, the rf excitation coil was designed to be “single-sided” by connecting the solenoidal rf coil in series with a planar coil which was inserted between the sample container and the oven [Fig. 1(a)]. The planar coil canceled the rf field generated by the solenoid in the region of the K vapor with a cancellation factor of about 80. Second, we employed a pulsed detuning field B_{off} along the z axis which shifted the K magnetic resonance frequency to about 2 MHz while rf pulses of $B_1 = 4.5$ G were applied. The offset field was also highly inhomogeneous, so that any transverse K polarization created by the rf pulse quickly dephased. At the end of a rf pulse, the offset field was slowly ramped down so that K spins adiabatically followed the net field $B_0 + B_{\text{off}}$. We verified that the magnetometer recovers its normal sensitivity within 0.8 ms after the termination of a rf pulse. In compari-

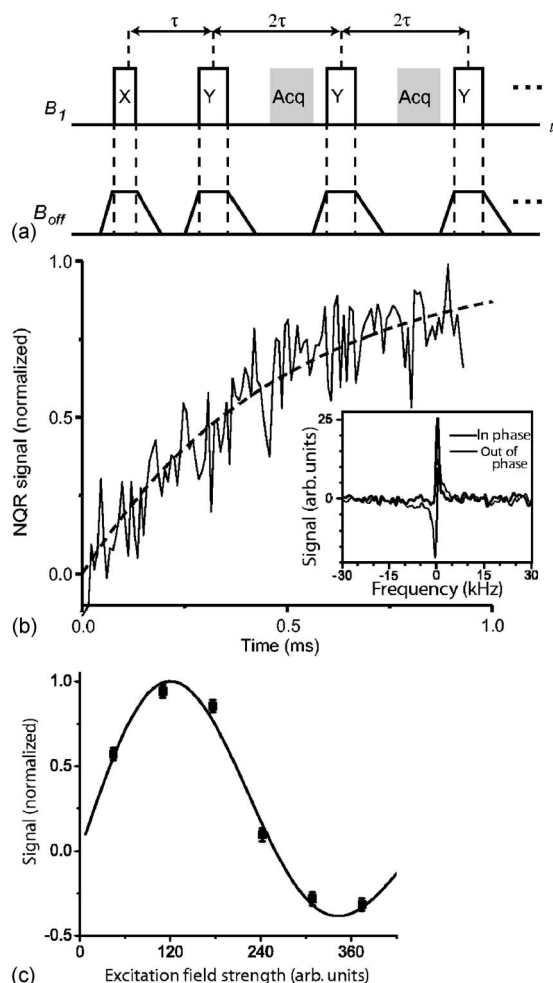


FIG. 3. (a) Timing diagram of the excitation $\theta=119^\circ_X$ and refocusing $\theta=150^\circ_Y$ rf pulses and offset field pulses. (b) Lock-in detected ¹⁴N NQR signal from powdered ammonium nitrate. The dashed line is a single exponential fit with a rise time of 0.5 ms. The inset shows a Fourier transform of the signal obtained in a different run, with total T_{1N} -limited acquisition time of 4 min. (c) The NQR signal amplitude as a function of the excitation field strength. The solid line is a fit to the function $J_{3/2}(\theta)/\theta^{1/2}$ following Ref. 11.

son, we observed that the magnetometer recovery time without the offset field was on the order of 30 ms, somewhat longer than the optical depth ($OD=62$, estimated from absorption spectrum) divided by the pumping rate ($\sim 7000 \text{ s}^{-1}$, calculated from the photon flux).

Because of the long $T_{1N}=16.6 \text{ s} \gg T_{2N}=4.4 \text{ ms}$ of ¹⁴N, the NQR signal was detected with a spin-lock spin-echo sequence¹² shown in Fig. 3(a). Here the initial excitation of the spins is followed by a series of refocusing pulses to form multiple spin echoes at intervals of $2\tau=2.2 \text{ ms}$. The magnetometer acquired signals during 1 ms acquisition windows between these pulses. The output of the polarimeter, after analog amplification, was detected in quadrature with a Tecmag Apollo console. Figure 3(b) shows the averaged echo signal after 32 repetitions of a 2048 echo sequence. The slow rise of the signal is due to the high Q of the magnetometer. The inset shows the Fourier spectrum of the signal after eight repetitions of the same sequence. In the inset figure the measured SNR of 25 corresponds to a SNR of 9 from a single such sequence that lasts for 4.4 s. Due to the long T_{1N} , the repetition rate was low, at about twice per minute. Figure 3(c) shows the signal amplitude as a function of the excita-

tion pulse amplitude, demonstrating good agreement with the predicted Bessel-function dependence.

In conclusion, we have demonstrated sensitive detection of a NQR signal from a room temperature solid using a newly constructed subfemtotesla atomic magnetometer. Among the three quantum-limited noise terms identified in Ref. 1, the photon shot noise term is found to be dominant in our setup due to relatively low probe beam intensity. Furthermore, excess noise from probe beam fluctuations raised the detected noise level to about twice that of the shot noise. With a more stable laser, and a moderate probe beam power of around 100 mW, a shot noise limited sensitivity of $\sim 0.06 \text{ fT/Hz}^{1/2}$ should be readily possible. As a comparison, we conducted a separate experiment of detecting NQR from NH_4NO_3 using a conventional, tuned-coil based probe with a loaded Q of 60 and a coil volume comparable to that of the magnetometer cell. The measured SNR corresponded to a field sensitivity of $3.6 \text{ fT/Hz}^{1/2}$ at 423 kHz, where the field refers to the average field over the coil. Following Ref. 13 the Johnson noise limited sensitivity of the same probe was calculated to be $0.8 \text{ fT/Hz}^{1/2}$. Note that even if a higher- Q coil could be constructed, it would improve the SNR relatively slowly as $\sim Q^{1/2}$.¹³ Ultimately, a coil with a very high $Q(\gg 1000)$ would limit the possibility of using a fast, multiple spin-echo sequence such as the one used here. We conclude, therefore, that the demonstrated sensitivity of the atomic magnetometer is significantly higher than that of a pickup coil probe, for samples that have to be placed outside the volume of the coil. In addition, an atomic magnetometer is much less susceptible to electronic interference because the detected NQR signal, converted to voltage by the photodiodes and transimpedance amplifiers, is on the order of a microvolt, whereas a typical NQR signal from a pickup coil is in the nanovolt range. Currently work is under way to realize sensitive detection in a more open environment, where external rf noise is canceled by subtracting signals from more than one probe beam. Such development will potentially lead to a mobile atomic NQR spectrometer for various field applications.

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