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Enhancement of the Two-Dimensional Conduction Electron Zeeman Energy Near $\nu = 1$ by Optical Dynamic Nuclear Polarization

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Enhancement of the Zeeman energy of 2D conduction electrons near $\nu = 1$ by optical dynamic nuclear polarization (DNP), as observed by the Overhauser shift of the transport detected electron spin resonance, is measured quantitatively for the first time in GaAs/AlGaAs multiquantum wells. The NMR signal enhancement is obtained under similar conditions in the same sample, allowing the hyperfine coupling constant of 3.7T between between the nuclei and 2D conduction electrons to be measured for the first time. The potential to suppress the Zeeman energy by optical DNP is discussed in the context of its potential influence on Skyrmion formation.

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A popular theme of recent studies of the quantum Hall effect is the characterization of nontrivial spin configurations which depend on the interplay between the Zeeman and Coulomb interactions of the two-dimensional electron system (2DES) [1–4]. Experimental investigations have provided strong support [5–11] for the now generally accepted theoretical prediction that charged spin texture excitations known as Skyrmions can occur in the vicinity of filling factors for which the Fermi level resides between levels with opposite electron spin. According to theory, the spatial extent of the Skyrmion is determined by the competition between the electron-electron Coulomb term Δ_0 and the Zeeman term $E_z = g\mu_B B_0$. Under conditions favorable for Skyrmion formation, excursions away from exact filling of a spin polarized level cause spin depolarization at a rate higher than would be expected for single spin-flips in a non-interacting many-electron system. A variety of techniques have been employed to measure the filling factor dependence of the 2DES spin polarization [5–7], $\langle S_z\rangle(\nu)$, in GaAs/Al$_x$Ga$_{1-x}$As heterostructures and multiquantum wells in the integer and fractional regimes, with most of the initial work focussed on $\nu = 1$. These techniques include: optically pumped NMR [5–7], thermally activated magnetoresistance in tilted fields [8], magnetic circular dichroism of optical transmission [9], heat capacity [10], and circular polarization of time-resolved recombination radiation [11].

Several methods are available to control the critical ratio E_z/Δ_0. The g-factor can be reduced by the application of hydrostatic pressure [12] or addition of aluminum into the quantum wall lattice [13]. However, the application of hydrostatic pressure or the introduction of aluminum results in a considerable reduction in the electron density and a severe degradation of the 2D electron mobility. Alternatively, the ratio can be controlled by the tilted field method [8,14–16], whereby the cyclotron energy, which depends only on the perpendicular component of B_0, is reduced by tilting the sample with respect to the field while E_z is unaffected. After rotating, B_0 is increased to return to the filling factor of interest, but with higher Zeeman energy. However, this method has several limitations: (*i*) it can only increase E_z/Δ_0, (*ii*) potentially introduces Landau level mixing effects and (*iii*) requires a high bandwidth tuning range in magnetic resonance experiments.

An ideal experiment would be one wherein $\langle S_z\rangle(\nu)$ can be measured as as function of E_z/Δ_0 at constant B_0. Toward this objective we introduce the notion that E_z can be enhanced or suppressed using the local nuclear hyperfine field, B_n, yielding $E_z = g\mu_B(B + B_n)$, where [17]

$$B_n = \frac{8\pi}{3}\frac{g_0}{g}\sum_j a^{(j)}\gamma_n^j|\psi(r^{(j)})|^2\langle I_z^{(j)}\rangle \tag{1}$$

is summed over all isotopes (i.e. ^{69}Ga , ^{71}Ga and ^{75}As), each with natural abundance $a^{(j)}$, gyromagnetic ratio $\gamma_n^{(j)}$, and nuclear spin expectation value $\langle I_z^{(j)}\rangle$ along the z-axis (Zeeman order). In the context of ESR, B_n is known as the Overhauser shift [18]. In the absence of the spin-orbit interaction, as in the conduction band of GaAs, neither the cyclotron energy nor the electron-electron Coulomb interactions are affected by B_n, regardless of its magnitude or sign, because the origin of B_n is the spin-spin coupling between the electron and nucleus. In semiconductor systems, $\langle I_z\rangle$ can be dramatically enhanced by microwave [17–19] or optical dynamic nuclear polarization (DNP) [20–26]. Enhancement of B_n by microwave or optical DNP has been previously reported in a variety of homogeneous and heterogeneous semiconductor systems. In bulk GaAs, optical DNP enhanced shifts causes the Hanle curve for depolarization of

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the shallow donor trapped electron-hole recombination photoluminescence to be displaced from zero field [27,28]. In $Al_xGa_{1-x}As/GaAs$ quantum dots, optical DNP enhanced effective nuclear fields of up to 1.0T have been observed in the spectrally resolved excitonic Zeeman splittings [29]. In a previous study of the 2DES in $Al_xGa_{1-x}As/GaAs$ heterostructures, magnetoresistance detection of ESR was used to measure microwave DNP induced Overhauser shifts as large as 430mT [30,31].

Here we report the first quantitative measurements of the enhancement of E_z for 2D conduction electrons near $\nu = 1$ by optical dynamic nuclear polarization (DNP), as observed by the Overhauser shift of the transport detected electron spin resonance in GaAs/AlGaAs multiquantum wells. The NMR signal enhancement is obtained under similar conditions in the same sample, allowing us to determine the hyperfine coupling constant between the nuclei and 2D conduction electrons to be measured for the first time. The factors governing the enhancement and potential suppression or E_z by optical DNP are discussed on the basis of the electron-nuclear scalar relaxation equation.

Using the technique developed by Stein et al. [32], we have detected ESR in the longitudinal magnetoresistance (ρ_{xx}) of three different $Al_xGa_{1-x}As/GaAs$ multiple quantum well samples, EA-124, EA-216 and EA-221, with 2D electron densities ranging from 7.0×10^{10} (EA-124) to $1.6 \times 10^{11}cm^{-2}$ (EA-221). The 4.2K mobilities are in the $5 \times 10^5 cm^2/V{\cdot}s$ range. Contacts to the 100μm wide Hall bar pattern were made using AuGeN. A second unpatterned sample from wafer EA-124 was set aside for the NMR experiments. We will concentrate on the results from EA-124 since the enhancement of $\langle I_z \rangle$ due to optical DNP was detected by both NMR and via the Overhauser shift in this sample under similar conditions. Sample EA-124 has a capping layer of 10nm GaAs and 100nm of $Al_{0.1}Ga_{0.9}As$, a quantum well widths of 30nm, and barrier widths of 360nm. The 2D electrons were introduced by Si δ-doping in the center of the barrier.

A YIG (yttrium indium garnet) oscillator with an output of $\approx$ 100mW over a range of $10 - 18$GHz and single frequency bandwidth of 1MHz served as the microwave source. A solid state doubling amplifier was employed to achieve $>$ 100mW microwave power in the $20 - 36$GHz range. For lockin detection of the $\Delta\rho_{xx}$ signal, the microwaves were amplitude modulated. The samples were rotated on a gear driven goniometer stage to obtain the ρ_{xx} minimum at $\nu = 1$ while maintaining the ESR frequency within the range of the microwave system. The microwave field was introduced through a coaxial transmission line terminated by a loop antenna. The ρ_{xx} and $\Delta\rho_{xx}$ signals were simultaneously recorded using an injection current of $1 - 3\mu$A. The ESR spectra were recorded by monitoring $\Delta\rho_{xx}$ at constant frequency while sweeping the external magnetic field at a typical rate of 3mT/s. The ESR data presented here were collected using either the Keck resistive magnet or using the 100mm 6T superconducting magnet at the National High Magnetic Field Laboratory. The ^{4}He bath temperature was determined from the vapor pressure and from a calibrated 100Ω carbon film resistor mounted 1.0cm above the sample.

The ESR data are summarized in Fig. 1. A g-factor of -0.41 was obtained in EA-124 while in the higher density samples EA-216 and EA-221 a slightly lower g-factor of -0.39 was measured. This decrease in magnitude of the g-factor with increasing magnetic field is in agreement with previous measurements in single heterostructures [33]. Note that the application of the modulated microwaves did not produce an appreciable change in ρ_{xx} detected at the current modulation frequency. The detection sensitivity is estimated to be 10^4spins/Gauss.

To observe the optical DNP enhanced Overhauser shift, the samples were illuminated through a 600μm diameter optical fiber mounted 1.0cm above the sample, resulting in an unpolarized light beam with a diameter of $\approx 4 - 5$mm. Figure 2 presents a plot of $B_n(t)$ in sample EA-124 as a function of the duration of exposure to unpolarized light. The exponential time dependence can be explained assuming that the mechanism of microwave or optical DNP in a 2DES is analogous to the mechanism involving electrons trapped at shallow donors in bulk GaAs. The scalar relaxation [17,30,25,26] of the non-equilibrium electrons via electron-nuclear flip-flops are induced by fluctuations in $A\mathbf{I}\cdot\mathbf{S}$. For short pumping times spin diffusion can be neglected and the time dependence obeys:

$$\frac{\langle I_z \rangle(t)}{I_0} = \left(1 - \frac{\gamma_e}{\gamma_n} S_0 \left(\langle S_z \rangle - S_0\right)\right) \left(1 + \frac{T_{IS}}{T_{1n}}\right)^{-1} \left\{1 - \exp\left[-t\left(T_{IS}^{-1} + T_{1n}^{-1}\right)\right]\right\} \quad (2)$$

where $1/T_{IS}$ is the electron-nuclear cross-relaxation rate and T_{1n} is the nuclear spin lattice relaxation time in the absence of light. This solution is independent of the statistics obeyed by the electrons and applies equally well for paramagnetic ions and electrons in a 2D metal [17,30]. The thermal equilibrium spin order of the electrons in the 2DES, S_0, is now filling factor dependent, but $S_0 \geq 0$ since $g < 0$. The ratio of the electron and nuclear gyromagnetic ratios, γ_e/γ_n, represents the enhancement of $\langle I_z \rangle$ over its thermal equilibrium value I_0. In the case of unpolarized light, $\langle S_z \rangle \rightarrow 0$ when equal numbers of $m_F = +1/2$ and $m_F = -1/2$ electrons are excited. Thus, unpolarized optical DNP is analogous to microwave DNP in the sense that the electron spin polarization is driven towards zero. Since $\gamma_S = \mu_B g_e < 0$ in GaAs and $\gamma_I > 0$ for all three isotopes, $\langle I_z \rangle < 0$ and hence $B_n > 0$. The effect can be viewed as a $(B + B_n)/B$ enhancement of the g-factor. The relaxation decays of B_n following unpolarized optical excitation and microwave excitation of EA-124 are also shown in Fig. 2. A maximum Overhauser shift of $B_n = +120$mT was observed using $\approx$ 50mW microwave power. The power at the sample is estimated to be on the order of a few mW.

Similar values for T_{1n} were obtained following optical and microwave DNP. It should be noted that these relaxation decays do not correspond to a single magnetic field (or filling factor) but rather to a T_{1n} averaged over the entire range of filling factors covered by the sweep.

The wavelength dependence of the B_n and the quadrupole split ^{71}Ga NMR transition amplitudes are presented in Fig 3. In the NMR sample the GaAs substrate was removed by chemical etching and the AlGaAs/GaAs multiquantum well film was epoxied to a Si substrate. Upon lowering the temperature a planar stress is induced due to a difference in thermal expansion between the film and the Si substrate. The NMR data were collected at a fixed field of 3T in a high homogeneity Oxford magnet. A typical optical DNP enhanced ^{71}Ga NMR spectrum is shown in the Fig. 3d. No Knight shift was observed at 4.2K. Three NMR lines, with FWHM$\approx$ 3kHz and separated by 55kHz are observed due to strain induced nuclear quadrupole interaction. Due to the $\approx 20\mu s$ dead time of the receiver it was necessary to extrapolate the complex free induction decays to the origin of time defined by the termination of the $1.5\mu s$ RF detection pulse. This procedure eliminated the linear phase shift. The NMR signal is predominantly due to nuclei in the GaAs quantum well regions. With longer optical pumping times the central peak is seen to grow disproportionately due to spin diffusion into the AlGaAs regions. The satellite transitions cannot spin diffuse into the barriers due to the mismatch in transition energy with the AlGaAs satellites which experience a substantial first order quadrupole broadening due to the presence of 10% aluminum [34]. This assignment is confirmed by comparing the known composition of the film with the thermal equilibrium NMR spectrum shown in Fig. 3e. The optical DNP enhancement is strongly wavelength dependent. The spectrum undergoes several 180° phase inversions. At 812.9nm a total phase inversion results when the laser wavelength is increased by only 0.335nm (150GHz). We propose that these inversions are due to spin state selective optical pumping in the lowest two Landau levels. Also note the antiphase relationship between the satellite and central peaks which is observed over the entire wavelength range. The form of the spectrum indicates that the density operator is dominated by a term corresponding to an octupole polarization. The distribution also appears to contain a residual dipole and quadrupole polarization. A full decomposition of this mixed multipole distribution could be obtained by recording the ratio of the double-quantum NMR transition intensities. The NMR phase inversions cease to occur at shorter wavelength, and simultaneously, the Overhauser shift abruptly increases.

The polarization of the central transition following a 40s optical excitation at 801.5nm is estimated to be $\langle I_z \rangle = 3.2 \times 10^{-3}$ which corresponds to the observed signal enhancement factor of 8.4 over the thermal equilibrium signal at $T = 2.4$K. Under similar optical DNP conditions, an Overhauser shift of $B_n = 6$mT is observed, which establishes $B_n = -3.7\langle I_z \rangle$. This is the first direct measurement of the collective hyperfine coupling constant between the conduction 2DES and quantum well nuclei. A larger value would result if it were not for the destructive interference between hyperfine field components resulting from the substantial octupole polarization. The measured value of the hyperfine coupling constant for this 2DES is remarkably close to the theoretical estimate of 3.53T for the shallow donor trapped electrons in bulk GaAs [22,30].

Finally, we discuss the possibility of suppressing the E_z of the 2D conduction electrons by optical DNP on the basis of the electron-nuclear scalar relaxation model. The enhancement is proportional to the deviation of the average electron spin from thermal equilibrium. In optical DNP, the steady state electron spin polarization $\langle \hat{S}_z \rangle$ is determined by the electric dipole transition matrix elements which govern the intensities of the direct interband transitions near the center of the Brillouin zone in a III-V semiconductor (GaAs type). This establishes $\langle \hat{S}_z \rangle = \mp 1/4$ for $\sigma^{\pm}$ (right or left circularly polarized) pumping light. The solution of the scalar relaxation equation predicts the magnitude and sign of $\langle I_z \rangle$ for arbitrary $\langle S_z \rangle$ and S_0. At sufficiently small values of S_0, such as at the even valued filling factors, optical orientation should provide the ability to obtain $-1/4 \leq \langle S_z \rangle \leq 1/4$. Hence, an increase or decrease of E_z can be achieved. Suppression of E_z should enhance Skyrmionic features in the fractional filling regime under practically attainable conditions of electron density, temperature and magnetic field. Optical DNP may be a useful method for enhancing or suppressing the Zeeman energy.

In summary, we have demonstrated that the effective Zeeman energy of a 2DES near $\nu = 1$ can be enhanced by optical DNP with unpolarized light. Although the maximum observed Overhauser shift was only 15mT, increased optical intensities, longer pumping times, and a judicious choice of the pumping field to minimize spin lattice relaxation is expected to increase this value by 1-2 orders of magnitude. These quantitative measurements of the Zeeman energy enhancement under optical pumping conditions may be valuable in NMR studies of $\langle S_z(\nu) \rangle$ under optical pumping conditions. [5–7]. Lastly, we have discussed the applicability of optical DNP suppression of the Zeeman energy as a method for enhancing Skyrmionic excitations in the quantum Hall effect.

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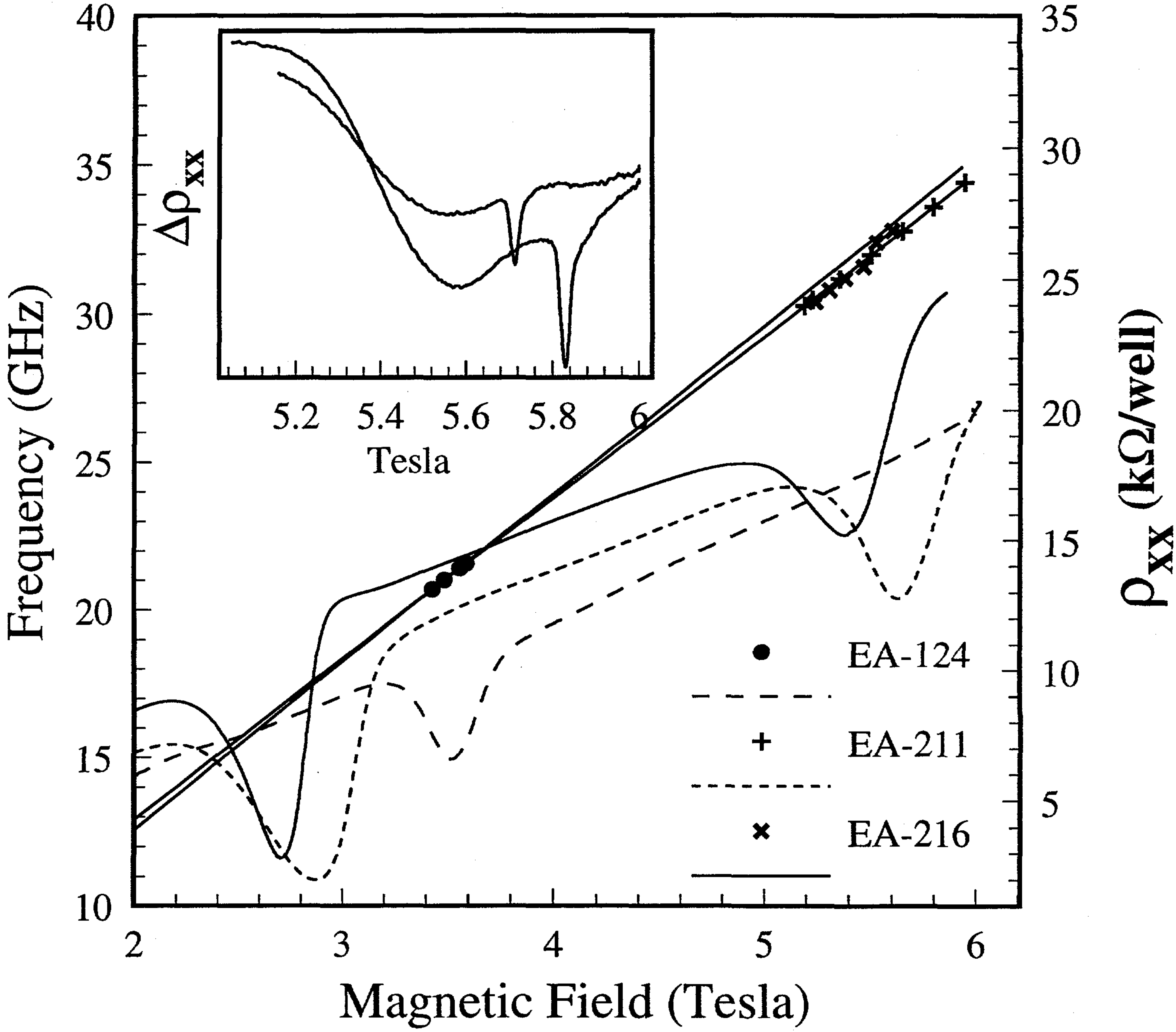
FIG. 1. Summary of transport detected ESR near $\nu = 1$, $T = 2.4$K in 3 different samples: EA-124, EA-211 and EA-216. Plots of ρ_{xx} correspond to the axis at right. The ESR responses at 2 microwave frequencies are shown in the inset.

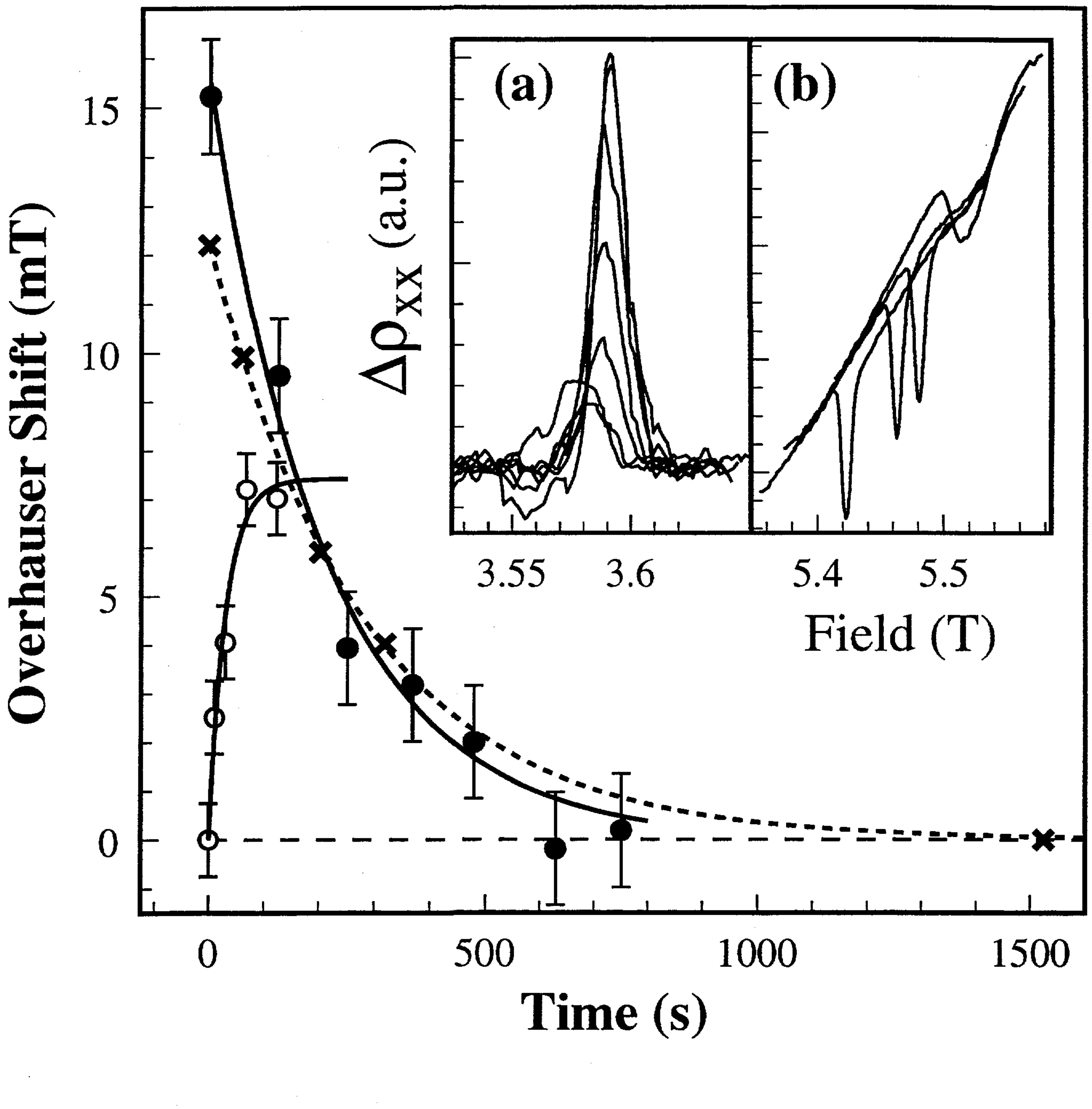
FIG. 2. Sample EA-124 data near $\nu = 1$. (a) ESR spectra, after non-resonant background subtraction, acquired following a 154s exposure at $\lambda = 784$nm and $I = 180\text{mW/cm}^2$. (b) ESR spectra following microwave DNP, as described in the text. In each case the nuclear relaxation was followed by repeated B_0 up-sweeps. Open circles: $B_n(t)$ pumping due to exposure to unpolarized light, with $\lambda = 800$nm and $I = 240\text{mW/cm}^2$, yielding a polarization time constant of 30.4s. Filled circles and crosses: relaxation decays of B_n following optical ($\lambda = 784$nm, $I = 180\text{mW}/cm^2$) and microwave DNP, yielding $T_{1n} = 214$s and $T_{1n} = 280$s, respectively. The microwave DNP induced Overhauser shift has been scaled down by a factor of 10.

FIG. 3. (a) Wavelength dependence of B_n enhancement in EA-124, $I = 106\text{mW/cm}^2$, $T = 2.4$K, $B_0 = 3.3$T. The wavelength dependence of the $|+1/2\rangle \rightarrow |-1/2\rangle$ and $|+3/2\rangle \rightarrow |+1/2\rangle$ NMR transition amplitudes are shown in (b,c), respectively, for sample EA-124 at $B_0 = 3.0$T, $T = 4.2$K, $I = 800\text{mW/cm}^2$. The inset (d) presents the Fourier transform NMR spectrum obtained using a 1.5μs resonant radio frequency pulse following a *40s* pumping period at $\lambda = 812.9$nm, $T = 4.2$K. The linear phase shift was removed in the time domain. Inset (e) shows the $T = 300$K ^{71}Ga NMR spectrum of the film after averaging 30,000 spectra.



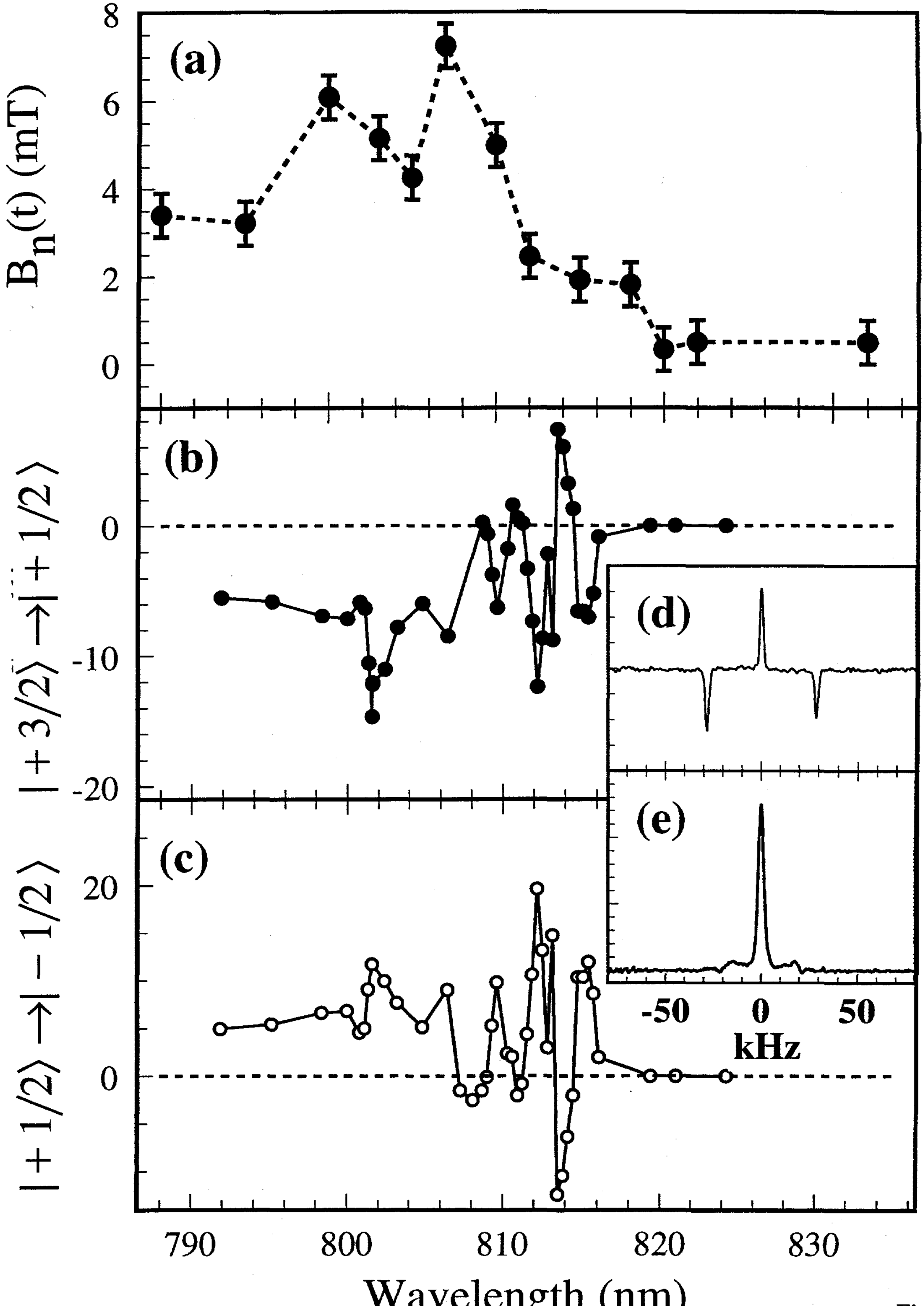
Frequency (GHz)
10
15
20
25
30
35
40
ρ_{xx} (kΩ/well)
5
10
15
20
25
30
35
2
3
4
5
6
Magnetic Field (Tesla)
$\Delta\rho_{xx}$
5.2
5.4
5.6
5.8
6
Tesla
EA-124
EA-211
EA-216

Fig. 1.



Overhauser Shift (mT)
15
10
5
0
0
500
1000
1500
Time (s)
(a)
(b)
$\Delta\rho_{xx}$ (a.u.)
3.55
3.6
5.4
5.5
Field (T)

Fig. 2.



(a)
(b)
(c)
(d)
(e)
$B_n(t)$ (mT)
$|+3/2\rangle \rightarrow |+1/2\rangle$
$|+1/2\rangle \rightarrow |-1/2\rangle$
8
6
4
2
0
-10
-20
20
-50
50
kHz
790
800
810
820
830
Wavelength (nm)

Fig. 3.