wave after the initial stage of damping[8] and the average level of its oscillation afterwards.

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[3]In order to apply the present formalism to the case of the pendulum, the rate of change of the length must be sufficiently small so that the Coriolis term can be neglected.

[4]Property (2) can be used to reduce (by half) complex analytical calculations based on electron motion, or it offers a powerful means of checking the results. I have verified its convenience in G. Pocobelli, Bull. Amer. Phys. Soc. 23, 794 (1978), paper 3S8.

[5]The failure of the adiabatic solution at the separatrix is quickly *reversed* under the present formalism, in which the phase can only grow with time and a matching set of solutions is produced naturally; (2) is avoided by using Fourier expansions of the Jacobian elliptic functions, which require only the always finite and continuous forms (3) and (3′).

[6]Equations (4) and (4′) represent the $\oint p dq$ adiabatic invariant.

[7]Pocobelli, Ref. 4.

[8]The damping of the wave is exponential (linear Landau damping) at the beginning.

Magic-Angle Line Narrowing in Optical Spectroscopy

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Spin decoupling and line narrowing are observed for the first time in an optical transition, ${}^3H_4 \leftrightarrow {}^1D_2$ of Pr^{3+} in LaF_3 at 2 °K, with use of optical free-induction decay. The ^{19}F nuclei, when irradiated by an appropriate rf field, undergo forced precession about an effective field at the magic angle in the rotating frame. The fluctuating ^{19}F-^{19}F dipolar interaction is thereby quenched and the optical linewidth drops from ~10 to ~2 kHz, as predicted in a theory of spin diffusion.

In the field of NMR, there exist several ways of reducing the time-dependent magnetic dipolar interaction between spins. Examples are motional narrowing,[1,2] macroscopic samples rotation,[3] spontaneous spin flip-flop processes,[4] and forced spin precession.[5-10] In this Letter, we report the first observation of this kind in an optical transition of a low-temperature solid, LaF_3:Pr^{3+}, where the dilute nuclear spin (I) is praseodymium and the abundant spin (S) is fluorine. The Pr^{3+} ions are coherently prepared by a laser field and thereafter exhibit nonlinear optical free-induction decay (FID). The Pr^{3+} dephasing time, as suggested in earlier work,[11] is limited by spin diffusion among the ^{19}F nuclei which undergo resonant flip-flops and impress weak fluctuating fields on the ^{141}Pr nuclei. This action adiabatically modulates the Pr^{3+} optical transition frequency through the dipolar I-S interaction and broadens the line. We now show that the half-width at half-maximum (HWHM) optical linewidth is reduced from ~10 to ~2 kHz when the fluorine spin diffusion process is quenched by application of suitable magnetic static and rf fields, causing the ^{19}F nuclei to precess about an effective magnetic field at the *magic angle* in the rotating frame. This observation enables us to identify in an unambiguous way that the ^{19}F-^{19}F dipolar interaction is the dominant optical line-broadneing mechanism and provides the first test of spin diffusion theory in an optical transition. As we shall see, the behavior at optical and rf frequencies is different.

Imagine that the Pr^{3+} ions are coherently prepared by a laser field in the optical transition $1 \leftrightarrow 2$ and then experience nonlinear FID when the laser frequency is switched outside the Pr^{3+} homogeneous linewidth. The novelty of this technique[11] is that a single homogeneous packet (≲10 kHz width) can be selected from the much broader inhomogeneous line shape (5 GHz width). The FID signal, expressed in terms of the induced polarization, is of the form

$$\langle p(t)\rangle = \langle p(0)\exp\{i[\omega_{12}t + \int_0^t \delta\omega_{12}(t')dt']\}\rangle, \quad (1)$$

where $\delta\omega_{12}(t')$ represents the fluctuation in the optical transition frequency ω_{12} due to the nuclear dipolar S-S and I-S interactions. The angular brackets denote an average over the optical inhomogeneous line shape, the geometric variables of the I-S interaction, and the S-S spin fluctuations. With the assumption of Markovian spin statistics, we apply the spin diffusion theory of Klauder and Anderson[12] and find that (1) predicts a Lorentzian homogeneous line shape having a HWHM linewidth

$$\Delta\nu = \left(\frac{4\pi\hbar}{9\sqrt{3}}\right) |(\gamma_I{''} I_z{''} - \gamma_I{'} I_z{'})\gamma_S S_z| n_S\left(\frac{r}{R}\right). \qquad (2)$$

Here, γ_I (~ 23 kHz/G) denotes the enhanced gyromagnetic ratio of $^{141}Pr^{3+}$ for the lower (double prime) or upper (single prime) electronic state. The fluorine spin S has a gyromagnetic ratio γ_S (4 kHz/G) and number density n_S, flips at the intrinsic rate r, and has a macroscopic rate parameter R, introduced by Klauder and Anderson[12] to assure stationarity.

Now consider the application of a static magnetic field B_0 and a radio frequency field $B(t) = 2B_x \cos\omega t$ which is detuned from the fluorine Larmor frequency $\gamma_S B_0$ by $\Delta_S = \gamma_S B_0 - \omega$. In a frame rotating at the frequency ω, the effective field $B_e = [B_x{}^2 + (\Delta_S/\gamma_S)^2]^{1/2}$ is stationary and makes an angle $\beta = \tan^{-1}(\gamma_S B_x/\Delta_S)$ with the static field B_0. When the ^{19}F precession frequency $\gamma_S B_e$ exceeds the square root of the S-S second moment, the ^{19}F precessional motion at the angle β will tend to be uninterrupted, and it is then advantageous to perform a transformation to a second rotating frame where the axis of quantization is parallel to $\vec{B}_e$.[8-10] In this tilted rotating frame, the secular part of the dipolar Hamiltonian $\mathcal{H} = \mathcal{H}_{SS} + \mathcal{H}_{IS}$ contains the time-independent terms

$$\mathcal{H}_{IS}(\beta) = \cos\beta\,\mathcal{H}_{IS}{}^{(0)}, \qquad (3a)$$

$$\mathcal{H}_{SS}(\beta) = \tfrac{1}{2}(3\cos^2\beta - 1)\mathcal{H}_{SS}{}^{(0)}, \qquad (3b)$$

where

$$\mathcal{H}_{IS}{}^{(0)} = \textstyle\sum_j 2(b_j{''} - b_j{'}) S_{jz} I_z$$

and

$$\mathcal{H}_{SS}{}^{(0)} = \sum_{k<j} a_{kj}(3S_{kz}S_{jz} - \vec{S}_k \cdot \vec{S}_j)$$

are the corresponding dipolar terms in the absence of an rf field, a_{kj} and b_j being the usual geometric factors.[8-10] We are now able to modify Eq. (2) by including the effect of an rf field on the Pr^{3+} optical linewidth. First, (3a) implies that $S_z(\beta) = \cos\beta S_z(0)$,[5] which replaces the heteronuclear term S_z in (2). Second, we associate the S-spin-flipping rate r in (2) with an inverse correlation time,

$$r(\beta) \equiv 1/\tau_c(\beta) = |\tfrac{1}{2}(3\cos^2\beta - 1)|/\tau_c(0),$$

derived previously by Mehring, Sinning, and Pines for NMR[9] with use of the result (3b). Equation (2) now takes the form

$$\Delta\nu(\beta) = \Delta\nu(0)|\cos\beta \cdot \tfrac{1}{2}(3\cos^2\beta - 1)|, \qquad (4)$$

and gives the Pr^{3+} optical linewidth as a function of the off-resonance angle β. Equation (4), which is shown in Fig. 1, predicts that the linewidth vanishes at the magic angle $\beta = \cos^{-1}(\frac{1}{3}\sqrt{3})$ or 54.7° and also at $\beta = \pi/2$, the fluorine resonance condition.

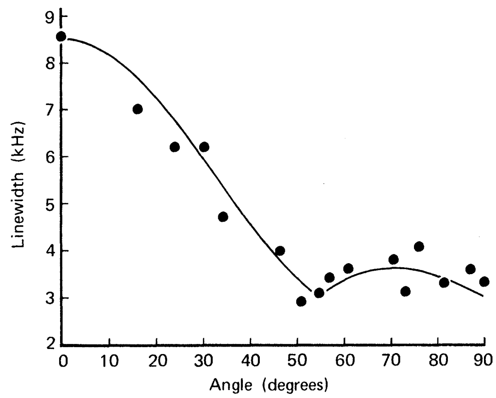

FIG. 1. Pr^{3+} optical linewidth vs the angle $\tan^{-1}(\gamma_S B_x/\Delta_S) = \beta$ expressed in degrees. Solid circles: experimental points for the case $B_0 = 130$ G$\|c$ axis and $B_x = 25$ G. Solid curve: Equation (4) with frequency offset of 3 kHz included for residual broadening.

The Pr^{3+} transition[11] examined, $^1D_2 \rightarrow {}^3H_4$ at 5925 Å, involves the lowest crystal-field components of each state. These are electronic singlet states that couple to the Pr^{3+} nuclear spin $I = \frac{5}{2}$ in second order, producing quadrupolar splittings of order 10 MHz and an enhanced nuclear gyromagnetic ratio γ_I. Three equally intense optical transitions occur, $I_z{''} \leftrightarrow I_z{'} = \pm\frac{5}{2} \leftrightarrow \frac{5}{2}$, $\pm\frac{3}{2} \leftrightarrow \pm\frac{3}{2}$, and $\pm\frac{1}{2} \leftrightarrow \pm\frac{1}{2}$, and overlap because of the large inhomogeneous strain broadening of ~ 5 GHz. We investigated the $\pm\frac{1}{2} \leftrightarrow \pm\frac{1}{2}$ transition, which is easily identified because it appears as a long decay following the faster $\frac{5}{2}$ and $\frac{3}{2}$ components, consistent with Eq. (2) and a computer simulation of a triexponential decay.

Optical FID is monitored by laser frequency switching[11] with use of the pulse sequence of Fig. 2. An acousto-optic Bragg modulator, driven by

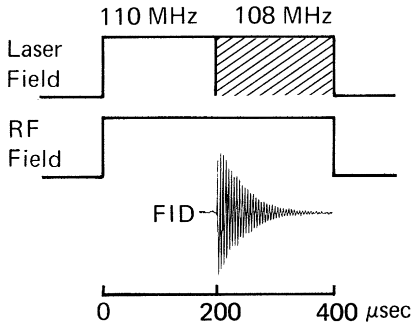

FIG. 2. Pulse sequence showing the laser field, its frequency shift, and the spin-decoupling radio-frequency field with time. The Pr^{3+} ions are coherently prepared by the laser field in the initial 200-μsec interval and then exhibit optical FID when the laser frequency is suddenly switched 2 MHz at $t = 200$ μsec.

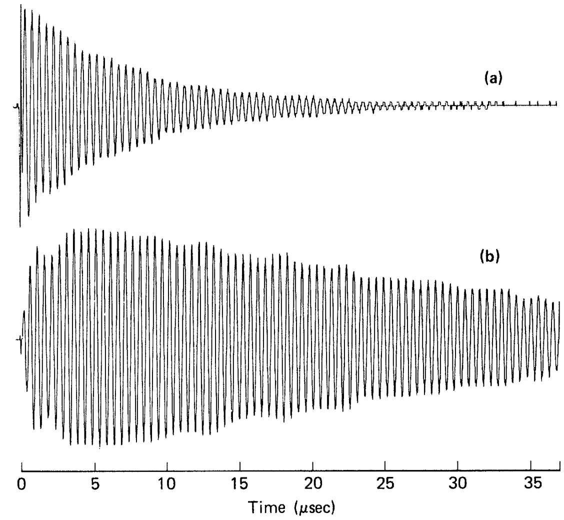

FIG. 3. Optical free-induction decay in LaF_3:Pr^{3+} at 1.8 °K in the presence of a static magnetic field $B_0 = 130$ G ⊥ c axis: (a) with no rf field, where $T_2 = 15.6$ μsec (10.2 kHz), and (b) with an rf field $B_x \approx 25$ G, where $T_2 = 66$ μsec (2.4 kHz).

a 110-MHz rf pulse generator, deviates the beam of a cw ring dye laser through a 1.5-mm aperture while imparting a 110-MHz laser frequency switch. Before and after the pulse the undeviated beam is blocked and since the pulse repetition rate is 1 Hz, complications due to optical pumping[11] are reduced. The transmitted beam propagates along the c axis (laboratory y axis) of a $5 \times 6 \times 7$ mm^3 crystal of LaF_3:Pr^{3+}(0.1% Pr^{3+}) before striking a p-i-n photodiode. The laser field is linearly polarized along the laboratory x axis and has a 1.0-mm diameter in the crystal at a power of 10 mW. In Fig. 2, we see that the Pr^{3+} ions are coherently prepared by the optical field during the initial 200 μsec of the pulse, and then FID occurs when the laser frequency shift is suddenly reduced from 110 to 108 MHz, the laser field remaining constant. Most important, broadening due to laser frequency jitter is reduced by frequency locking the ring laser to a passive external reference cavity, the laser stability being ~1 kHz over the duration of the measurement, ~50 μsec.

For the spin decoupling experiment, an rf coil oriented along the x axis is in close contact with the crystal and provides a pulse with a rotating component $B_x = 25$ G which is variable over the frequency range 0 to 600 kHz and is time coincident with the optical pulse of 400 μsec duration (Fig. 2). Both coil and crystal are immersed in liquid helium at 1.8 °K. A static magnetic field $B_0 = 130$ G is oriented along either the z or y laboratory axis (⊥ or ∥ to the crystal c axis) and exceeds the local field so that the nonsecular dipolar terms are small. Individual FID signals, which appear at a 2.003-MHz beat frequency because of heterodyne detection with the laser field, are captured with a Biomation model 8100 transient recorder for reproduction on an X-Y chart recorder (Fig. 3). In the absence of power broadening, the observed dephasing time is $T_2/2$.

A clear demonstration of optical line narrowing by spin decoupling is indicated in Fig. 3 for the case $B_0 \perp c$ axis. In the absence of rf, trace (a), the linewidth at HWHM is 10.2 kHz ($T_2 = 15.6$ μsec). In the presence of rf, trace (b), the value is 2.4 KHz ($T_2 = 66$ μsec), where the rf frequency $\omega/2\pi = 450$ kHz corresponds to the magic angle 54.7°. Additional confirmation is obtained by varying the rf frequency over the range from off resonance, $\beta = 0$, to on resonance, $\omega/2\pi = 520$ kHz or $\beta = \pi/2$. Figure 1 shows that the observed optical linewidth for the case $B_0 \parallel c$ axis follows the theoretical expectation Eq. (4), where a frequency offset of 3 kHz is added to account for residual broadening. Furthermore, at frequencies above resonance ($\beta > \pi/2$), the experimental pattern repeats with the mirror symmetry predicted by (4). For the case $B_0 \perp c$ axis, the behavior is similar but not identical to Fig. 1 because of the Pr^{3+} hyperfine anisotropy. Our theoretical model for spin decoupling therefore is confirmed in some detail and clearly exposes the magnetic origin of the optical line-broadening mechanism.

On the other hand, spin decoupling in NMR has revealed different characteristics, partly because the FID observed in the rf region is a first-order process and thus both dynamic and static interactions contribute to the linewidth. In systems such as AgF, the F spin diffusion process can motionally narrow the NMR ^{109}Ag resonance,[4] and when the spin diffusion process is suppressed as it is at the magic angle, the linewidth *broadens* rather than narrows.[9,10] Line narrowing[9,10] is observed, however, at the F resonance condition $\beta = \pi/2$.

The the magic angle, the optical linewidth of ~2 kHz appears to be limited by residual laser frequency jitter. Of course, a fundamental limit of 160 Hz is set by a $^1D_2$ radiative lifetime of 0.5 msec. We estimate that at 1.8 °K the phonon broadening linewidth is only 7 Hz, and in the preparative FID stage, laser power broadening and the effect of a finite optical pulse width of 200 μsec are negligible. As the laser frequency stability is improved further, it will be possible to examine weaker interactions which otherwise would be obscured in the absence of spin decoupling, an example being the ^{141}Pr-^{141}Pr interaction. Thus, a new family of spin decoupling experiments can be carried out for the first time at optical frequencies, allowing the manipulation and study of the basic dynamic processes.

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Visual Observation of the Second Characteristic Mode in a Quasiperiodic Flow

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The flow between concentric cylinders with the inner cylinder rotating has been studied by visualization techniques. Velocity power spectra have shown that the flow is described by two characteristic frequencies before it becomes chaotic; the first frequency corresponds to traveling azimuthal waves. The second frequency is identified as a modulation of the waves and multiple values have been observed. These are believed to be the first such observations for any nonlinear nonequilibrium system.

Recent studies of instabilities in convecting fluids[1] and in flows between rotating cylinders[2,3] and spheres[4] have shown that these systems, in addition to possessing a periodic regime characterized by a single frequency f_1, also often exhibit quasiperiodic behavior characterized by a second frequency f_2 which is in general incommensurable with the first.[5] Presumably such quasiperiodic behavior is common to a large variety of nonlinear nonequilibrium systems.[6] While the spatial character of the mode with frequency f_1 is well understood, no identification has been made of motion which might correspond to the frequency f_2, and no mathematical or physical relationship has been established between f_1 and f_2. Such information would aid significantly in the development of realistic mathematical models of these systems.

Using flow visualization techniques we have observed a wave motion that corresponds to the second characteristic frequency in the velocity power spectrum of a fluid contained between concentric cylinders with the inner cylinder rotating (circular couette flow). At small Reynolds number R, proportional to the angular velocity of the inner cylinder, the flow is purely azimuthal, but