of the incident electrons is doubled, that of the channeling radiation increases at least as fast: The sharp peaks in the planar-channeling spectrum increase by slightly more than a factor of 3 and the broad enhancement in the axial-channeling spectrum increases by about a factor of 2. From these data one can infer that, in this energy range, the scaling for the planar case is somewhat slower than  $\gamma^2$ , and that for the axial case is about  $\gamma$ .

The largest enhancements relative to bremsstrahlung were observed for axial alignments, but the nearly identical "shoulder" spectrum of Fig. 3(d) casts some doubt upon a strict interpretation of the axial enhancement as channeling radiation alone. In contrast with the axial case, planar features occur only within small angles of the channeling direction. Also for the planar case, the enhancements relative to random bremsstrahlung are approximately 2:1 at 56 MeV. Since the estimated effective coherence lengths are  $\simeq 1 \ \mu m$ , these enhancement ratios can be expected to improve by more than an order of magnitude if a 1- $\mu$ m-thick crystal were used.

In conclusion, we have observed peaks in the low-energy emission spectrum from electrons channeled in silicon. The estimated planar coherence lengths indicate that very large enhancements relative to ordinary bremsstrahlung should be achieved with thinner crystals. The existence of unpredicted spectral structure establishes that the enhancement results from correlated motion (channeling) of the electrons over a considerable distance ( $\geq 1000$  Å) in the lattice, and requires that a successful description of the effect must involve a quantum-mechanical treatment.

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## Ultrahigh-Resolution Spectroscopy: Photon Echoes in YA1O<sub>3</sub>:Pr<sup>3+</sup> and LaF<sub>3</sub>:Pr<sup>3+</sup>

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By using a new, delayed-heterodyne photon-echo technique, we have obtained a spectral resolving power of  $1.2 \times 10^{11}$ , measuring a 2.0-kHz linewidth (half width at half maximum) for the 6105-Å  ${}^{1}D_{2} - {}^{3}H_{4}$  transition of YAlO<sub>3</sub>:Pr<sup>3+</sup> in an external magnetic field. This appears to be the highest resolution ever obtained in optical spectroscopy. Measurements were also made on the corresponding transition of  $LaF_3$ :  $Pr^{3+}$ giving a linewidth of 5 kHz. Optical free induction decay gave incorrect results (i.e., significantly broader linewidths).

The field of optical spectroscopy has witnessed a dramatic improvement in resolution since the advent of highly stable lasers, and of techniques

for eliminating the effects of inhomogeneous broadening (i.e., photon echo,<sup>1</sup> optical free induction decay,<sup>2</sup> fluorescence line narrowing,<sup>3</sup> and

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saturation spectroscopy or hole burning<sup>4</sup>). In this Letter we report the measurement of a homogeneous linewidth of 2.0-kHz half width at half maximum (HWHM) for the 6105-Å  ${}^{1}D_{2} \rightarrow {}^{3}H_{4}$  transition in YAlO<sub>3</sub>:Pr<sup>3+</sup>, using a delayed-heterodyne-photon-echo technique. This corresponds to a spectral resolving power or line  $Q [= \nu/2\Delta\nu(\text{HWHM})]$  of  $1.2 \times 10^{11}$ , to our knowledge the highest resolution yet obtained in optical spectroscopy.

In the quest for ultrahigh resolution, two limiting factors intervene. The first is the intrinsic homogeneous linewidth of the optical transition, and the second is the frequency stability of the laser being used to perform the measurement. The first limit can be lowered by selecting the appropriate material and transition. Previously the highest-resolution optical spectra have been obtained in gas-phase systems:  $Q = 4.4 \times 10^{10}$  in CH<sub>4</sub> by Hall, Bordé, and Uehara<sup>5</sup> and  $Q = 8 \times 10^{10}$ in Ca by Barger  $et al.^6$  In these systems there is a special position in the inhomogeneous line that can be accurately selected, viz. the group of atoms or molecules with zero velcity. This makes them useful for absolute frequency standards. In solids where inhomogeneous broadening is due to random crystal strains this is no longer true, and they cannot be used as frequency standards. In other respects, however (e.g., absence of transit-time broadening), they are attractive candidates for high-resolution spectroscopy. Weak *f*-*f* transitions in dilute samples of rareearth ions in a low-temperature host exhibit negligible thermal broadening and the radiative lifetimes are very long (typically ~  $10^{-3}$  sec). In order for the optical linewidth to approach the limit set by the population decay rate, however, one must reduce or eliminate dephasing due to hyperfine coupling between the rare-earth and host atoms. We chose the system  $YAlO_3$ :  $Pr^{3+}$  because the  $Pr^{3+}$  ion has no first-order magnetic moment and is surrounded by oxygen atoms which have no nuclear spin. Hence, as pointed out recently by Erickson,<sup>7</sup> one expects the fluctuating hyperfine fields (from the more distance <sup>27</sup>Al and <sup>89</sup>Y) to be rather small.

Resolution limitations due to the laser were overcome by a combination of techniques. First of all, the frequency jitter of the cw dye laser was reduced from ~ 1 MHz to ~ 10 kHz on a microsecond time scale by a fast servo lock, in the manner of DeVoe *et al.*<sup>8</sup> The remaining jitter can be effectively eliminated by photon-echo experiments in which the system is coherently excited by a  $\frac{1}{2}\pi$ - $\pi$  pulse sequence. The second pulse, at

time  $\tau$ , reverses the inhomogeneous dephasing, producing a burst of radiation at  $2\tau$ ,<sup>1</sup> and the decay of the echo amplitude as a function of the delay time  $2\tau$  is the Fourier transform of the spectrum of the excited atoms. Thus, in these timedomain measurements, spectral linewidths are given by the echo decay and very small splittings can be resolved by observation of echo-amplitude modulation.<sup>9,10</sup> In the time domain there is no sharp resonance as a function of laser frequency ---such resonances appear as long echo-decay times. With photon echoes the laser needs to be stable only during the two pulses, which are much shorter than the dephasing time  $T_2$ . Furthermore, even laser jitter during the pulses is irrelevant as long as it remains small compared with the spectral width of the exciting pulses which is much greater than  $T_2^{-1}$ . This is in sharp contrast to optical free induction decay (FID) or saturation spectroscopy experiments, where the laser must remain stable to much less than a homogeneous linewidth for the duration of the experiment (i.e., at least  $T_2$  for FID, or the scan time for saturation spectroscopy). In our case the laser jitter of a few kilohertz was much less than the  $\sim 1$ -MHz bandwidth excited by the pulses so that we were able to eliminate laser jitter from our linewidth measurements. We also note that the echo decay is not affected by power broadening, whereas care must be taken in FID measurements that  $\chi^2 T_1 T_2 \ll 1$ , where  $\chi$  is the Rabi flopping frequency.<sup>2</sup>

Our experiments were performed on a sample of YAlO<sub>3</sub>:0.1%  $Pr^{3+}$  in liquid helium at 1.9 K. About 10 mW of 6105-Å light was focused onto the sample with an 85-mm-focal-length lens and propagated down the crystal *c* axis.<sup>7</sup> The laser was frequency stabilized by locking to an external Fabry-Perot cavity of free spectral range 7.5 GHz, and using an intracavity electro-optic phase modulator crystal to apply high-frequency corrections via a fast servo loop.<sup>11</sup>

The two-pulse echo sequence was obtained by amplitude gating the laser with an acousto-optic modulator having rise and fall times of 50 nsec. The echo amplitude was maximized by  $\frac{1}{2}\pi$  pulses of 2 µsec which prepared a bandwidth of ~ 1 MHz inhomogeneous line. Although a much smaller number of ions is excited than in broadband, high-power pulse preparation, the loss is recovered by heterodyne detection of the echo, in the following way: The optical frequency of the  $\frac{1}{2}\pi$ and  $\pi$  pulses is shifted with respect to the laser by 80 MHz, the rf used to drive the acousto-optic

modulator. Hence the echo frequency also contains an 80-MHz shift. Just before the echo appears, a 76-MHz rf pulse is applied to the modulator, producing a frequency-shifted light beam which gives a 4-MHz heterodyne beat echo signal [see Fig. 1(a)]. This technique combines the advantages of amplitude gating<sup>1</sup> and Stark<sup>12</sup> or frequency switching.<sup>13</sup> Heterodyne detection increases the size of the echo signal by  $10^2 - 10^3$  and also shifts its frequency from dc to several megahertz so that lower-frequency laser-amplitude variations can be easily filtered out. Although the difference between our delayed heterodyne technique and frequency switching<sup>12, 13</sup> may seem slight, it is very important to delay the onset of the heterodyne pulse as echo decays measured with the heterodyne source applied at the end of the  $\pi$  pulse gave (power-dependent) decays up to a factor of 3 times faster. The cause of this large effect is currently under investigation. A similar intensity-dependent dephasing has been observed in Stark-switched echo measurements on CH<sub>3</sub>F.<sup>14</sup> An advantage of the gated cw echo configuration is that the spectral width excited by the preparation pulses can be much less than the hyperfine splittings, thus avoiding the complication of deeply modulated echo decays.<sup>9</sup> However, because these splittings are much less than the inhomogeneous optical linewidth, different hyperfine transitions are excited (for different ions) and contribute to the observed echo decays. With somewhat more powerful cw lasers the length of the preparation pulse could be varied to measure specific splittings via echo modulation.<sup>10</sup> Single-shot echo signals were recorded on a transient recorder with good signal to noise [Fig. 1(a)]. The amplitudes of five shots were recorded at each pulse separation  $\tau$ . In contrast to FID measurements (see below) data taken in this way are very reproducible.

As in the case<sup>15</sup> of LaF<sub>3</sub>:Pr<sup>3+</sup>, hole burning occurs due to optical pumping of the nuclear hyperfine levels.<sup>7</sup> One way of eliminating this is to scan the laser slowly<sup>16</sup> (0.5 kHz/µsec). Another method which we found works well is to eliminate hole burning by saturating the two quadrupole transitions in the ground state with simultaneous irradiation at 7.05 and 14.1 MHz. In order to saturate all transitions in a magnetic field ( $\vec{H}_0 \parallel c$ ), sidebands produced by frequency modulating the 7-MHz rf source at the  $M_s = \pm \frac{1}{2}$  Zeeman frequency were added. This gives the same echo decay times as scanning the laser, provided that the rf transition rate is much less than the echo decay



FIG. 1. (a) Photon echo observed with the delayedheterodyne technique: 1 is the  $\frac{1}{2}\pi$  pulse, 2 is the  $\pi$ pulse, and 3 marks the beginning of the heterodyne pulse. (b) Photon-echo decays recorded at 1.9 K (each point being the average of five shots). Linewidths are HWHM.

rate.

Photon-echo decay curves for the 6105-Å  ${}^{3}H_{4}$ -<sup>1</sup> $D_2$  transition of YAlO<sub>3</sub>:0.1% Pr<sup>3+</sup> were recorded at 1.9 K for external magnetic fields  $(H_0)$  between 0 and 100 G. In zero field the dephasing time was found to be 35  $\mu$ sec (4.5±0.2 kHz HWHM). This was obtained from a least-squares average of five runs, a typical one being shown in Fig. 1(b). The linewidth decreased rapidly with applied field, saturating at a value of  $2.0 \pm 0.2$  kHz for fields above 40 G. To show that the 2.0-kHz width was intrinsic, and not limited by laser jitter, we repeated the measurements using the internal lock on the CR599 (~1-2 MHz jitter) and obtained essentially the same width, i.e., 2.2 kHz. However the shot-to-shot reproducibility was much poorer.

To ascertain the relative sensitivity of photon echo and FID measurements to laser jitter, we measured single-shot FID decays using acoustooptic frequency switching and heterodyne detection.<sup>8</sup> Since the laser jitter is comparable to the homogeneous linewidth, very few (~ 1%) of the decays escaped modulation due to laser-frequency jumps during preparation. The narrowest FID widths obtained were 11 kHz ( $H_0 = 0$ ) and 5.6 kHz  $(H_0 = 80 \text{ G}; \text{ see Fig. 2})$ . This indicates that several kilohertz of laser-jitter contribution remains in the FID measurements.

Because of the intense current interest in  $LaF_3:Pr^{3+}$ ,<sup>8, 10, 15, 16</sup> we also measured photon-echo decays in this system using the  ${}^1D_2 \rightarrow {}^3H_4$  transition at 5925 Å. For  $H_0 = 0$  we obtained a homogeneous width of 28 kHz HWHM; for  $H_0 = 80$  G,  $\tilde{H}_0 \perp c$ , and at 7 kHz; and for  $H_0 = 80$  G,  $\tilde{H}_0 \parallel c$ , and at 6 kHz. These values are ~4 kHz less than we measured in corresponding FID decays. They are also significantly narrower than the widths recently obtained by DeVoe *et al.*<sup>8</sup> from FID, *viz*. 44 kHz ( $H_0 = 0$ ) and 10 kHz ( $H_0 = 76$  G,  $\tilde{H}_0 \perp c$ ), or those of a number of earlier measurements.<sup>15-17</sup>

The origin of the remaining 2-kHz linewidth in  $YAlO_3$ :  $Pr^{3+}$  is clearly of interest. The contribution of population decay ( $T_1$ ) processes was determined from a simple fluorescence-decay measurement which gave  $T_1 = 160 \ \mu$ sec in reasonable agreement with Erickson's value of 185  $\mu$ sec.<sup>7</sup> Thus the  $T_1$  contribution is 0.5 kHz HWHM. The remaining 1.5 kHz may be due to either residual hyperfine broadening from <sup>27</sup>Al and <sup>89</sup>Y nuclear spin fluctuations, or  $Pr^{3+}-Pr^{3+}$  interactions which have been found to contribute tens of kilohertz at this level of concentration in the  ${}^{3}P_{0}$  transition of LaF<sub>3</sub>:Pr<sup>3+</sup>.<sup>10</sup> Further measurements at lower concentrations are planned.

The hyperfine contribution to the homogeneous



FIG. 2. (a) Optical free induction decay measured at 1.9 K with  $H_0 = 80$  G,  $\mathbf{H}_0 \parallel c$ . (b) FID curves with and without an applied magnetic field  $H_0$ . Linewidths are HWHM.

optical linewidth in both YAlO<sub>3</sub>:Pr<sup>3+</sup> and LaF<sub>3</sub> :Pr<sup>3+</sup> is determined by the effect on ground and excited states of the fluctuating part of the hyperfine fields from neighboring ions. As expected, the linewidths obtained in LaF<sub>3</sub> are broader than those in YAlO<sub>3</sub> because of the presence of the near-neighbor F<sup>-</sup> ions. The reduction in homogeneous width in YAlO<sub>3</sub> (by a factor of 6) follows roughly the ratio of inhomogeneous nuclear-quadrupole-resonance linewidths (4:1).<sup>7, 15</sup> Although these linewidths are due to the static part of the hyperfine fields acting on the ground state of the Pr<sup>3+</sup> ion, they give an estimate of the relative coupling strengths.

We believe that the effect of the magnetic field is to modify the interactions among the nuclear spins. This can act to decouple <sup>27</sup>Al or <sup>19</sup>F nuclei from the Pr<sup>3+</sup> ions so that the average fluctuating hyperfine field at the  $Pr^{3+}$  site is substantially reduced. This has been reported in ruby, for example,<sup>1, 9, 18</sup> one difference being that there is a large electronic magnetic moment on the  $Cr^{3+}$ ions in ruby, and the field required to align the neighboring <sup>27</sup>Al nuclei is several kilogauss.<sup>18</sup> In addition, the echo decay will be affected by the rate of fluctuation of the local fields<sup>19</sup> which is also modified by the presence of the external magnetic field. The magnetic field dependence of the decays cannot be due to a modification of the optical pumping cycle<sup>8</sup> since elimination of optical pumping by the saturating rf fields did not affect the measured width.

In conclusion, we have shown that the delayedheterodyne photon-echo technique is well suited to measuring very long dephasing times. FID measurements, on the other hand, can give misleading results due to greater sensitivity to laser-frequency jitter, power broadening, and intensity-dependent dephasing arising from the presence of the heterodyne field. The latter can also be important in Stark- or frequency-switched echo measurements but is eliminated in the present technique. The resolution we have achieved in the  ${}^{3}\!H_{4} \leftrightarrow {}^{1}\!D_{2}$  transition of YAlO<sub>3</sub>: Pr<sup>3+</sup> in an 80-G field, i.e., a line Q of  $1.2 \times 10^{11}$ , appears to be the highest yet obtained in optical spectroscopy, and is comparable to that of Mössbauer spectroscopy (e.g.,  $Q \approx 10^{12}$  for Fe<sup>56</sup>).<sup>20</sup>

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## Superbanana Orbits in Stellarator Geometries

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The presence of superbanana orbits localized to either the interior or the exterior of classical stellarators and torsatrons is investigated for 3.5-MeV  $\alpha$  particles. The absence of the interior superbanana in both geometries is found to be related to nonconservation of the action. Exterior superbananas are found in the stellarator only, as a consequence of the existence of absolute localized magnetic wells. No superbananas of either type are found in the torsatron.

Trapping of charged particles in magnetic field ripples is an important consideration in studies of particle and energy transport properties of stellarators and torsatrons.<sup>1</sup> Superbanana trapping is one of the most harmful types of trapping. A superbanana is defined here as a trapped particle which is localized to a limited region of the torus, such that its excursions are restricted in both the toroidal and poloidal directions to some fraction of 360°. These localized particles make large radial excursions across the flux surfaces. enhancing the transport. In early numerical studies,<sup>2,3</sup> superbananas localized to the exterior of the torus were computed for a stellarator geometry. A later theoretical treatment of neoclassical transport in stellarators<sup>4</sup> derived a type of superbanana localized to the interior of the torus, as a consequence of the assumed invariance of the action.

In this Letter we consider the conditions under which both types of superbananas might occur in classical stellarators and torsatrons. We also study the invariants of motion in both geometries, using orbits computed from the Lorentz-force equation of motion. Particular attention is given to the action and its assumed invariance. This assumption is central to current neoclassical transport theory for these devices.

We begin with a review of the derived superbanana orbit, and find a range of magnetic moments for which these orbits should occur in a given configuration. We then test for superbananas with an independent numerical calculation, using exact fields for a corresponding filamentary configuration.

In the analytic derivation for the superbanana's orbit which follows, the magnetic field is assumed to follow the stellarator expansion for