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PSEUDO-STARK EFFECT AND FM/STARK DOUBLE
MODULATION SPECTROSCOPY FOR THE DETECTION
OF STATISTICAL FINE STRUCTURE IN ALEXANDRITE

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ABSTRACT: Using laser-FM spectroscopy and transient spectral hole-burning, the R_{1m} transition of Cr^{3+} ions in alexandrite is shown to exhibit a linear pseudo-Stark effect with coefficients of 0.141 MHz-cm-V^{-1} and 0.0538 MHz-cm-V^{-1} at 1.6 K for electric fields along the crystal a and c axes, respectively. This result was used to develop a sensitive FM/Stark double modulation method for the detection of statistical fine structure (SFS) on the inhomogeneously broadened R_{1m} line. While the final signal-to-noise ratio is not much greater than unity, autocorrelation of the measured spectra provides the first strong evidence for the presence of SFS in an inorganic material.

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I. Introduction

Recently, we reported the first observation of statistical fine structure (SFS) on an inhomogeneously broadened absorption profile [1,2] using the $S_1 \leftarrow S_0$ zero-phonon transition of pentacene in p-terphenyl mixed crystals and the technique of laser frequency-modulation (FM) spectroscopy. [3] SFS is time-independent structure on the inhomogeneous line caused by fundamental statistical variations (number fluctuations) in the spectral density of absorbers in each frequency interval. Observation of SFS in the spectra of pentacene in p-terphenyl was facilitated by the large low temperature absorption cross-section ($\approx 10^{-10}$ cm^2) and the very small persistent hole-burning quantum yield. [2] In many ways this system is ideal for the task of observing SFS, and many other organic mixed crystal systems where the guest has high oscillator strength and hole-burning is weak will have similarly favorable properties. However, in order to illustrate the generality of the SFS phenomenon, we report in this paper the results of our measurements on a transition metal ion (with low oscillator strength) in an inorganic host crystal.

The system chosen for this study is the lowest energy electronic absorption of Cr^{3+} ions in alexandrite (chrysoberyl) crystals. As we will show, this system has very different properties than pentacene in p-terphenyl, and presents challenging experimental problems in the observation of SFS. The principal problems are that the small oscillator strength in alexandrite ($\approx 10^{-5}$ compared to ≈ 0.1 for pentacene) leads to small absolute absorption changes due to SFS, and that the alexandrite excited state is easily saturated due to the relatively long excited state lifetime. We chose to detect SFS using a combination of FM spectroscopy and Stark modulation, and since the pseudo-Stark effect for the electronic transitions in alexandrite has not been previously reported, it was necessary to perform some preliminary studies on the electric field behavior of the spectra. We report here our measurements of the pseudo-Stark effect on transient holes in the spectra of alexandrite and its use as a method of performing double-modulation spectroscopy for the detection of SFS.

II. Experimental

The alexandrite ($BeAl_2O_4$:Cr^{3+}) crystal structure[4] is orthorhombic *Pnma* with four molecules per unit cell. There are two crystallographically inequivalent sites occupied by Al^{3+} ions in the lattice: one site having inversion (C_i) symmetry and the other having mirror (C_s) symmetry, with the mirror along the crystal *ac* plane. The Cr^{3+} ions substitute for Al^{3+} ions with about 78% going into the mirror site.[5,6] The alexandrite crystal used in all experiments was a highly polished nearly cubic single crystal about 5 mm on a side, with faces cut parallel to the crystallographic axes. The Cr^{3+} doping level was ≈ 0.03 atom % Cr/Al.

The Cr^{3+} transition studied in this work was the lower energy component of the $^2E_g \leftarrow {}^4A_{2g}$ transition for the mirror site ions, which is split by the distortion to C_s symmetry and is labeled the R_{1m} line.[7,8] At 1.6 K this zero-phonon line is centered at ≈ 679.62 nm (vacuum wavelength measured by a Burleigh WA-10 wavemeter) with a full-width at half-maximum (FWHM) of ≈ 2.2 cm^{-1} and is strongly *b*-axis polarized with a peak optical density greater than 4 at this Cr^{3+} concentration. In all experiments, the laser polarization was maintained along the *b*-axis and the laser wavelength was located on the long-wavelength edge of the line.

Laser FM spectroscopy was implemented in the manner described fully in Reference 2. Figure 1 shows a simplified apparatus diagram: the output of a single-frequency cw dye laser DL (Coherent 599-21, DCM dye) was sent through an electro-optic phase modulator EO (Lasermetrics 1097, $LiTaO_3$) driven by an rf oscillator and amplifier at 148 MHz, using up to 16 V rms into 50 Ω. This resulted in modulation indices between 0.3 and 1. The laser beam was first expanded and then focused by a 350 mm focal length lens to give a $\approx$ 65μm diameter spot throughout the entire 5 mm length of the sample. The sample was held between polished stainless steel electrodes in a liquid helium immersion cryostat, C. The transmitted beam intensity was measured by a Si avalanche photodiode-preamplifier

combination whose output was sent to a double-balanced mixer M referenced to the rf oscillator. The mixer output near dc was then either applied to the input of a lock-in amplifier (Princeton Applied Research HR-8) whose output was digitized and averaged by a digital scope DS (Data Precision 6100 with a 610 plug-in) for double-modulation detection, or sent to DS directly for ordinary laser-FM detection. In the double-modulation experiments, a modulated Stark field perpendicular to the propagation direction of the laser beam was applied to the sample using a unipolar square wave of up to 400 V at 10 kHz. This was accomplished using a high voltage power supply and an optically coupled high voltage switcher, HV. A signal synchronous to the high voltage modulation was used as a reference for lock-in detection. In this way, only those components of the laser-FM signal which were modulated at 10 kHz were detected. Not shown in the diagram are provisions for switching the FM sidebands and Stark modulation on and off as necessary in synchrony with the 0.25 s laser scan.

Transient holes were burned and probed by controlling the dye laser scan externally in the following manner. The laser was held at a fixed frequency a few hundred MHz from one end of the scan range for about 200 ms with the FM sidebands and Stark modulation switched off. This process burned a single hole in the spectrum, uncomplicated by pseudo-Stark splittings or holes burned by the FM sidebands. The laser was then rapidly scanned to the near end of its range, the FM and Stark modulations were turned on, and then the laser was scanned continuously to the opposite end of its range in 250 ms while digitizing the signal. After this, the laser was scanned rapidly back to the hole frequency, and a new hole was burned. This sequence was carried out repetitively and successive scans were averaged by DS. The total scan range was typically 2.5 GHz. No evidence for persistent hole-burning was observed under any conditions.

For the measurement of SFS spectra (as opposed to the measurement of transient holes), the laser was scanned forward and backward smoothly and continuously across its range with

the Stark and FM modulations on all the time. This was done to reduce interfering signals from the turn-on of the FM sidebands and to prevent the formation of transient holes within the scan range. Even so, weak transient holes are produced when the laser frequency is stationary for a brief time at the limits of the scan. To prevent these holes from distorting the SFS spectra, only the central half of the scan range was digitized and analyzed. The total scan range in these experiments was typically 1.0 GHz.

III. Results and Discussion

A. Pseudo-Stark effect

The ground state pseudo-Stark effect[9] for Cr^{3+} ions in the mirror site of alexandrite has been reported previously in ESR spectra at room temperature.[10] We demonstrate here that the R_{1m} line exhibits a linear pseudo-Stark effect when an electric field is applied along the crystal *a* or *c* axes. This is a pseudo-Stark effect, because there are two inequivalent mirror sites in an electric field, one of which shifts in one direction with applied field and the other in the opposite direction. As has been shown before for other zero-phonon transitions, the Stark effect can be precisely measured at high resolution using hole-burning techniques.[11,12] Figure 2 shows the results of a Stark experiment with several values for the applied (dc) Stark field E_s, with spectra measured by standard FM spectroscopy (the signal from only one FM sideband is shown, and the dc Stark field was turned on only during hole scanning). These spectra were obtained with $\vec{E}_s \parallel c$; similar results were obtained for $\vec{E}_s \parallel a$. Each spectrum is the average of 64 scans. Trace 2a shows the transient spectral hole with $\vec{E}_s = 0$ burned at 679.679 nm ($\equiv 0$ GHz) at 1.6 K with about 6.5 μW on the sample. Traces 2b-2f were similarly obtained using applied voltages of 50 V, 100 V, 200 V, 300 V, and 400 V, respectively. As the figure shows, the hole splits symmetrically into two components, each with approximately the same width as the unshifted line. We have plotted these shifts as a function of the applied field to determine the pseudo-Stark coefficients for $\vec{E}_s$ applied along

each of the three axes, as shown in Figure 3. Circles, squares and triangles correspond to applying $\vec{E}_s$ along the a, b and c axes, respectively. No effect was seen for the case of $\vec{E}_s \parallel b$, as expected for the $\Lambda'' \to \Lambda'$ transition of a 3d ion at a site of C_s symmetry. The straight lines were obtained from least-squares fits to the points, clearly demonstrating a linear dependence on field. The resulting pseudo-Stark coefficients for the shift of a single component are 0.141 MHz-cm-V^{-1} and 0.0538 MHz-cm-V^{-1} for $\vec{E}_s$ along the a and c axes, respectively. These coefficients are similar to those for ruby[11], as expected.

B. FM detection and transient hole behavior

Figure 4a shows a typical trace for laser-FM only detection of a transient hole burned at 1.6 K into the R_{1m} line at 679.684 nm ($\equiv$ 0 MHz) using $\approx$ 7.5 μW laser power. For this experiment, the Stark field was left off. The features at $\pm$ 148 MHz are the FM signals of the transient hole using cosine (F_1) detection,[2] and the spacing between these replicas of the hole allows calibration of of the frequency scale to 1 part in 10^6. The large transient at the left edge of the trace is due to a shift of the mixer dc output level caused by dc residual amplitude modulation[13] (dc-RAM) due to the FM sidebands being turned on at this point in time. The other, smaller features (which are most obvious between the two signals from the transient hole) are due to frequency-dependent RAM. This hole was power broadened considerably due to the conditions chosen for this experiment, and exhibits a width of 14.8 MHz (*vide infra*). Since the hole is transient and therefore constantly decaying, the two replicas have different intensities; only $\approx$ 60% of the hole intensity seen by the leading sideband is left at the time when the trailing sideband scans the hole. The time difference between when the passage of the two sidebands through the hole is 32.7 ms which, together with the observed hole decay and assuming a single-exponential decay process, implies a lifetime of $\approx$ 35 ms. This value is much greater than the 2.3 ms low-temperature lifetime of the ^2E_g state as measured by fluorescence decay.[7, 8] However, since the ground state is degenerate with a splitting[14] of 0.54 cm^{-1}, it is reasonable to expect that there might be

ground state cross-relaxation processes occurring which increase the hole recovery time, as has been observed for transient holes burned in the R_1 line of ruby. [15] No significant difference in width was observed for the two FM components of a transient hole, implying there are no important spectral diffusion processes occurring on a time scale faster than several tens of milliseconds.

Although it is not the principal goal of this paper to provide a careful measurement of the homogeneous width for the R_{1m} transition, at lower laser intensities we did observe shallow transient holes whose widths were ≈ 4.6 MHz, implying an upper limit to the zero-field homogeneous linewidth of one-half of this value, or 2.3 MHz. Since this width is comparable to the laser jitter linewidth for a 250 ms period, we assume that the actual homogeneous width is smaller. The only other report of the homogeneous width at zero field for Cr^{3+} in alexandrite[16] is 44 MHz, as determined by fluorescence line-narrowing techniques. The difference in the two measurements may be due to the fact that the crystal used in our measurements is more dilute (0.03 atom % versus 0.05 atom %).

C. FM/Stark double modulation spectroscopy

The purpose of laser-FM/Stark double modulation spectroscopy is to achieve true zero-background conditions in order to detect the very weak spectral features due to SFS. Without double modulation, the sensitivity threshold of FM spectroscopy can be limited to features greater than 10^{-4} in absorbance change due to RAM effects in the modulator. [17] The approach here is to modulate the spectral features at some low frequency v_s far from the FM modulating frequency, and detect the v_s component of the mixer output using lock-in detection. In this way, none of the RAM background is detected, allowing an increase in the gain of DS without overload from spurious background signals.

Trace 4b shows a simulation of double modulation detection of a hole similar to the one shown in Trace 4a, using a Stark shift appropriate for an applied field of 50 V along the

a-axis. In double modulation detection, the LIA sees an unshifted hole in one detection half-cycle, and two pseudo-Stark-split holes in the other detection half-cycle. When the LIA rectifies the signal at the reference input frequency, these two half-cycles are, in effect, subtracted from each other, resulting in lineshapes similar to those in Trace 4b. This is seen in the measured data in Figure 4c, which was obtained using $E_s = 50$ V along the a-axis. Note that the trailing sideband signal at +148 MHz is shallower as expected. For this value of E_s, the Stark lines strongly overlap the unshifted line (Stark shift = 14.4 MHz, linewidth = 14.8 MHz). At higher values of applied field, the Stark components shift farther from the unshifted hole position, and the signal increases because the oppositely signed components no longer overlap. As can be seen by comparing traces 4a and 4c, the double modulation technique very efficiently suppresses the RAM background seen using simple FM detection, effectively increasing the ultimate detection limit.[17]

D. SFS measurements

Acquisition of SFS spectra in alexandrite is complicated by several factors. First of all, the signal is expected to be small (10^{-4} in absorbance change). Usually the laser power is increased in order to lower the relative contribution of shot noise and improve the sensitivity to weak signals, but in the present case the laser power had to be kept below a few μW in order to prevent power broadening. This is the major difference between the present FM measurements and previous studies[17] in gases in which several mW of power could be presented to the detector. The long lifetime of the transient holes which may be burned in the spectrum also interferes in the following way. As the laser carrier frequency and the two sidebands sweep the spectrum, each component excites some of the ions into the excited state. Because of the lifetime of the transient hole and the scan rates and modulation frequencies used, the trailing sideband sees a spectrum which has been changed by the passage of the leading sideband and the carrier center frequency. More importantly, this "perturbed" spectrum can in principle be *different* for each laser scan, unless the exact same

ions undergo excitation by the leading sideband and center frequency every time the spectrum is scanned, which is unlikely. If such a process is occurring, then successive averages of many scans can be different, even in the absence of spectral diffusion or persistent hole-burning and trenching which might cause permanent changes in the "true" shape of the SFS. Also, if ground or excited state cross-relaxation is occurring, then it is possible that the "true" SFS spectrum changes after each scan. These effects can be minimized somewhat by lowering the laser power, but then the overall detection signal-to-noise would degrade.

Figure 5 shows typical results for attempts to detect SFS in the R_{1m} line of alexandrite at 1.6 K using laser-FM/Stark double modulation. Trace 5a shows two superimposed acquisitions of the SFS spectrum, each of which is the average of 4096 laser scans (0 GHz $\equiv$ 679.676 nm). The spectra are plotted in units of $(\Delta\alpha)L$, where α is the absorption coefficient, L is the sample length and $(\Delta\alpha)L$ is a measure of the difference in absorption between the two FM sidebands. One can see that there are obvious correlations of both the narrow and broad features of these two spectra, but even with the large number of averages taken the signal-to-noise ratio is near unity. Shown in trace 5b is a typical spectrum obtained when the laser is tuned off the R_{1m} line, again with 4096 averages taken with constant laser power at the detector. Due to the large noise background from laser shot noise and detector avalanche noise and to the effects mentioned above, the presence of SFS is not convincing when observing the raw spectra.

More convincing evidence for the presence of SFS structure is obtained by computing the normalized autocorrelations[1,2] of the spectra. These are shown in Figure 6, where traces 6a and 6b correspond to the spectra in 5a, and trace 6c corresponds to 5b. The autocorrelations of spectra taken with the laser in resonance with the R_{1m} line show a significantly broader origin feature than those taken with the laser off-resonance, suggesting the presence of underlying homogeneous lineshapes recurring in the on-line spectra. This broadening is most likely due to SFS, with the width of the origin feature reflecting the width

of the absorption features producing the SFS.[1,2] The width at the origin of the autocorrelation of off-resonance spectra is determined by the bandwidth of the detection electronics together with the digitization rate of DS. In the previous reports of SFS, the widths at the origin of (on-resonance) autocorrelated spectra were determined principally by the underlying homogeneous linewidth of the features making up the inhomogeneous band.[1,2] Here however, the width at the origin for traces 6a and 6b is comparable to the short-time laser jitter linewidth, $\approx$ 1-2 MHz. We therefore interpret our data as showing that the zero (magnetic) field homogeneous width of the Cr^{3+} ions in the R_{1m} line of alexandrite is less than 2 MHz, limited in our experiment by the laser linewidth. This result is consistent with the transient hole widths reported in Sect. III.B. above.

IV. Conclusion

Using laser-FM spectroscopy and transient spectral hole-burning, we have measured the Stark coefficients for Cr^{3+} ions in alexandrite for laser light polarized along the b-axis and find a linear pseudo-Stark splitting for the mirror site ions. Using this information, we have developed a FM/Stark double modulation detection technique that removes the interfering background due to residual amplitude modulation. We have used this technique and autocorrelation analysis to provide strong evidence for the existence of statistical fine structure in the R_{1m} transition of alexandrite at 1.6 K. This is the first evidence for SFS in an inorganic material and suggests that SFS will be a general feature of all inhomogeneously broadened transitions. In order to improve the signal-to-noise ratio of SFS measurements on weakly allowed transitions where excite-state saturation may be a problem, a detection technique is required that will provide higher laser power on the detector while maintaining the laser power on the sample below levels that produce power broadening. One technique that may achieve this is heterodyne FM spectroscopy, where the optical carrier is produced by a separate, higher power beam that does not pass through the sample.

ACKNOWLEDGEMENT

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FIGURE CAPTIONS

Figure 1. Simplified apparatus diagram. See text for description of symbols. A detailed description of the FM apparatus is presented in Ref. 2.

Figure 2. Pseudo-Stark effect on transient holes burned into the R_{1m} line of alexandrite at 1.6K, detected using laser-FM spectroscopy. The signal from only one FM sideband is shown here. Traces a) through f) correspond to applied voltages of 0, 50, 100, 200, 300 and 400 V, respectively.

Figure 3. Pseudo-Stark splittings as a function of applied electric field. The laser polarization is always along the crystal b axis. Circles, squares, and triangles correspond to applied electric fields along the a,c and b axes, respectively. The straight lines are least-square fits to the data. No splitting was observed for electric fields applied along the b axis.

Figure 4. Transient hole burning and laser-FM/Stark double modulation detection in the R_{1m} line of alexandrite at 1.6K. Trace a) is the signal from a transient hole detected with laser-FM spectroscopy; trace b) is a simulation of the lineshape expected for double modulation detection; trace c) is the actual double modulation signal of a transient hole obtained using identical conditions to those used to produce trace a).

Figure 5. SFS in alexandrite at 1.6 K using double modulation detection with a 10 kHz signal at 200 V applied along the a axis. Trace a) shows the superimposed results of successive averages of 4096 laser scans across a small portion of the R_{1m} line. Trace b) is an average of 4096 scans, with the laser tuned off of the line.

Figure 6. Normalized autocorrelations of the SFS data presented in Figure 5. Traces a) and b) were obtained from the on-line SFS spectra, trace c) was obtained from the off-line spectrum.
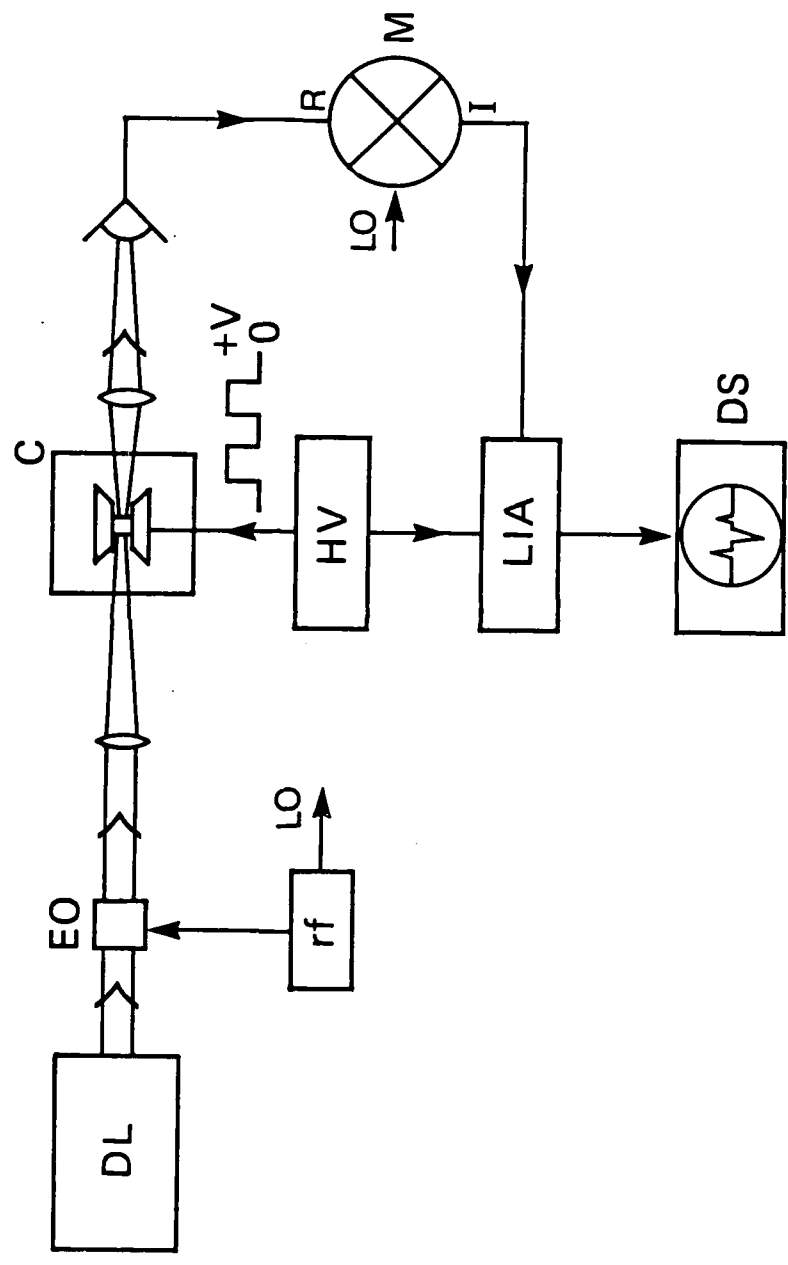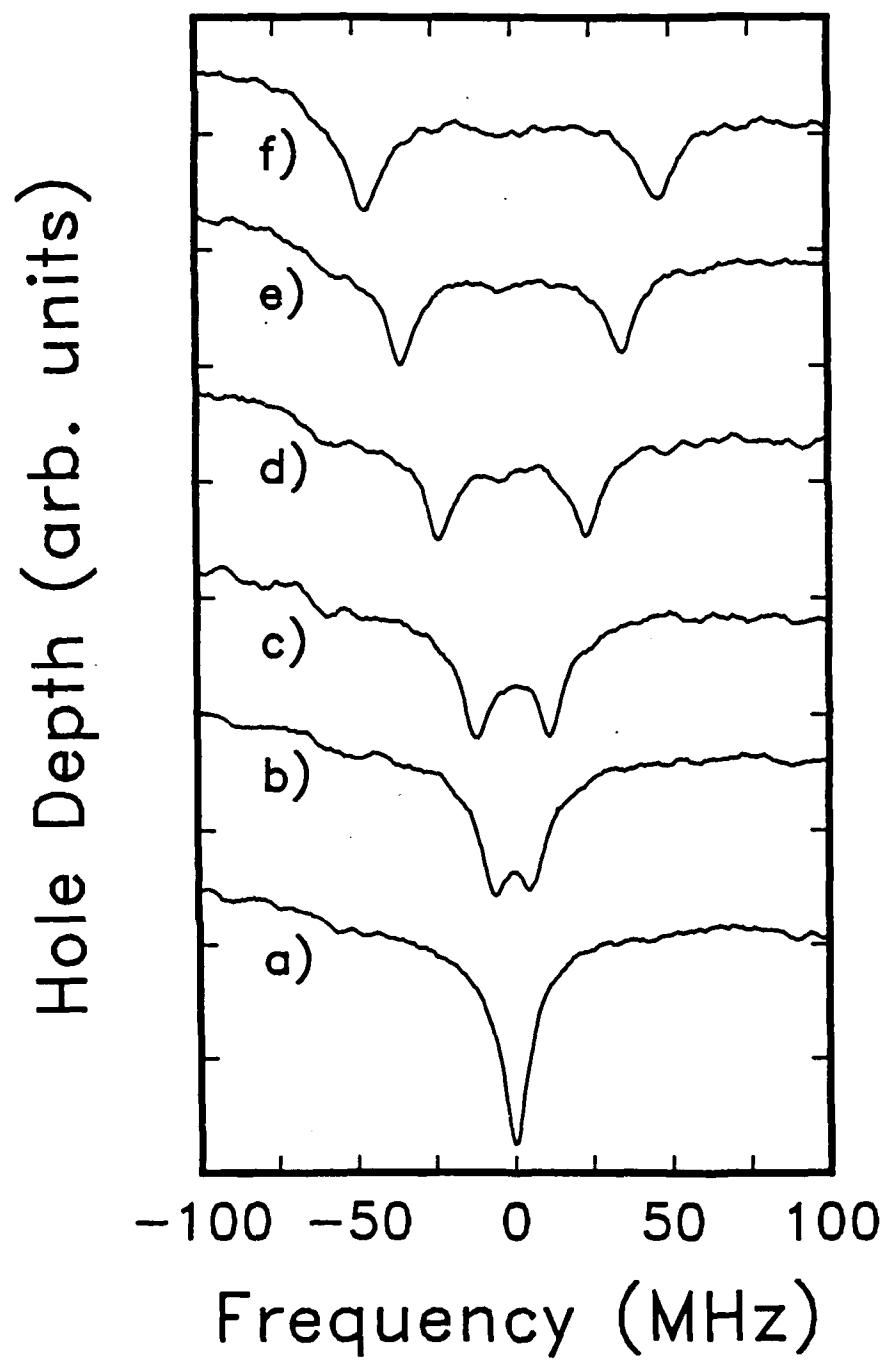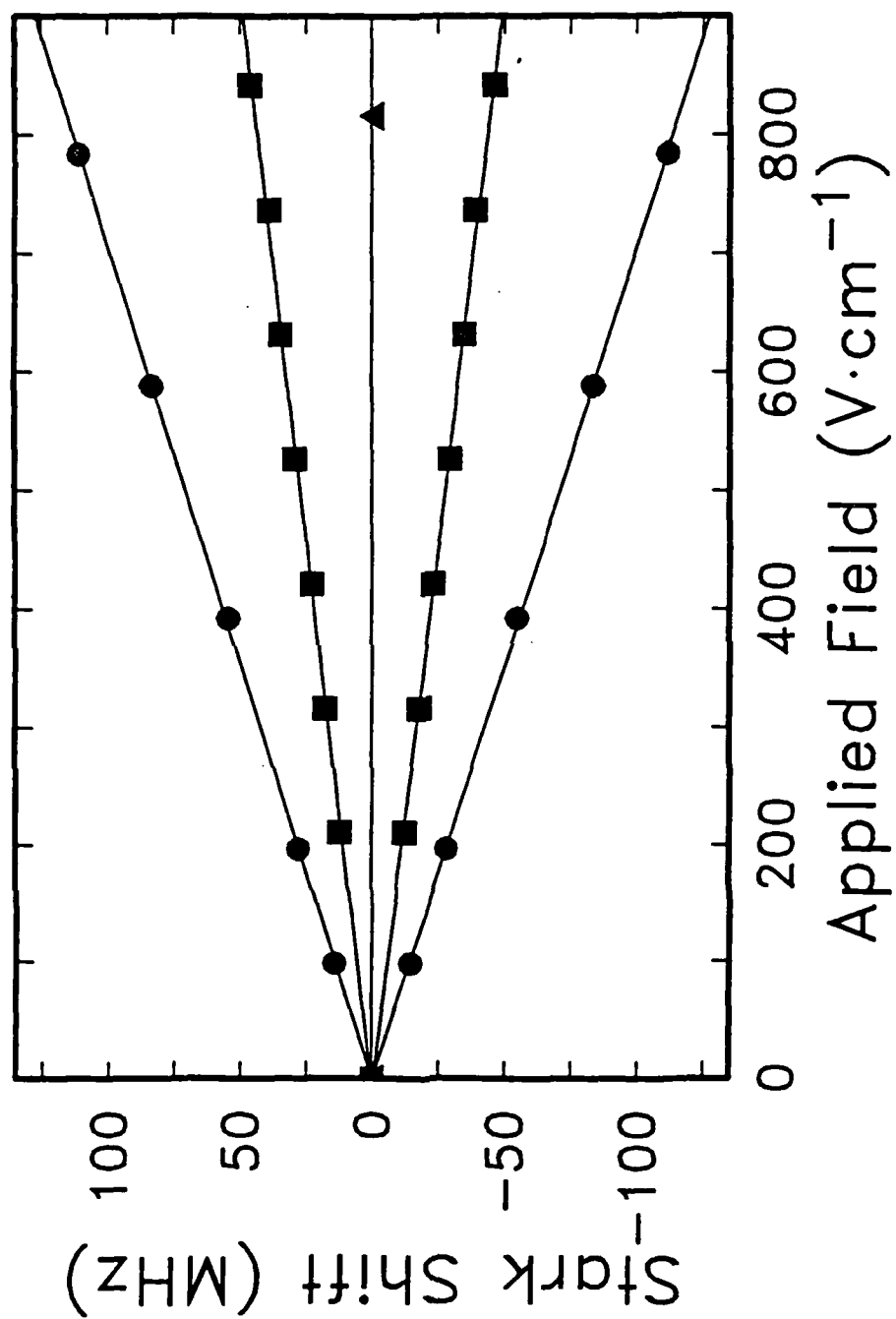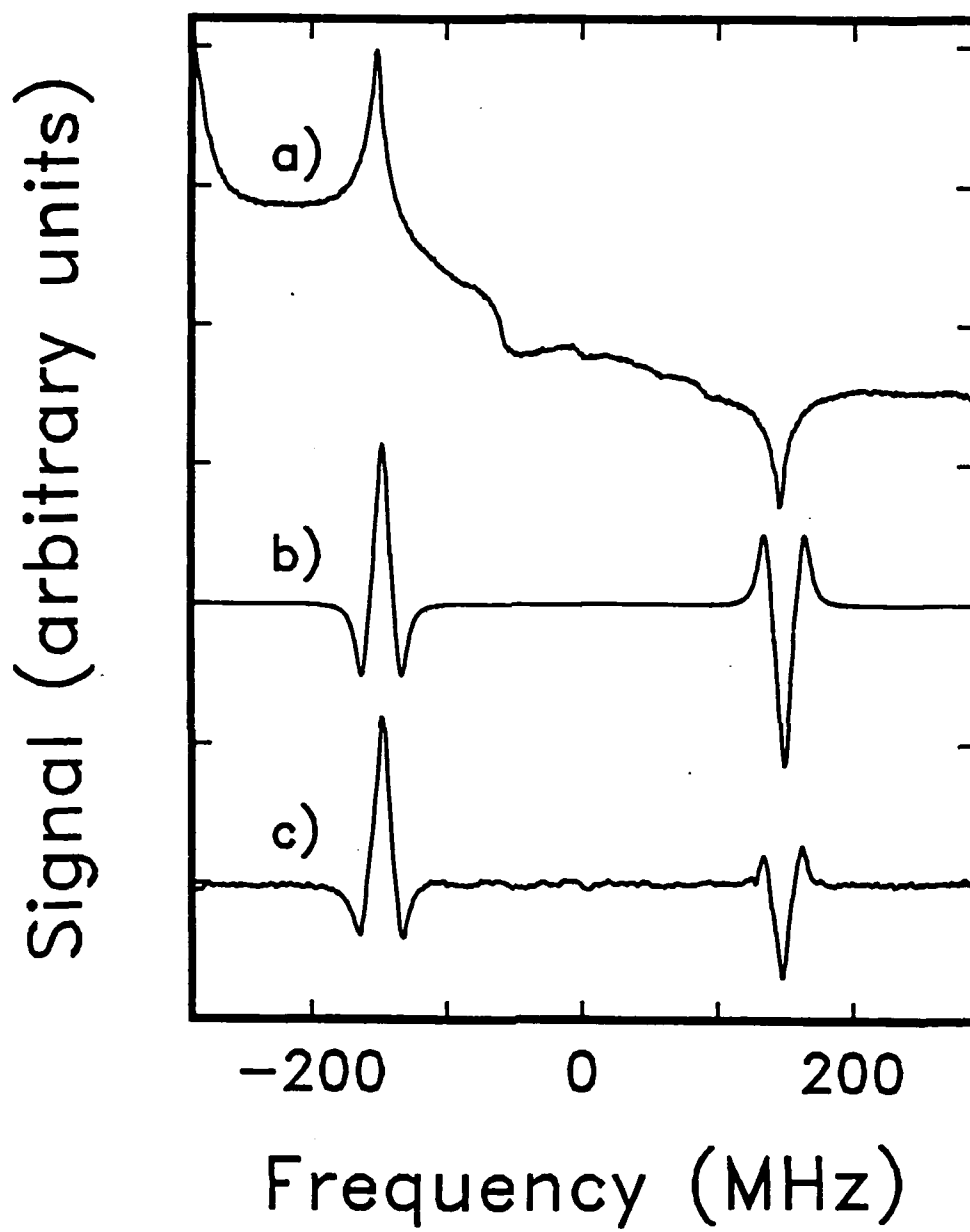
Figure 1

Figure 2

Figure 3

Figure 4

y-axis: $10^4 (\Delta\alpha)L$

x-axis: Frequency (MHz)

a)

b)
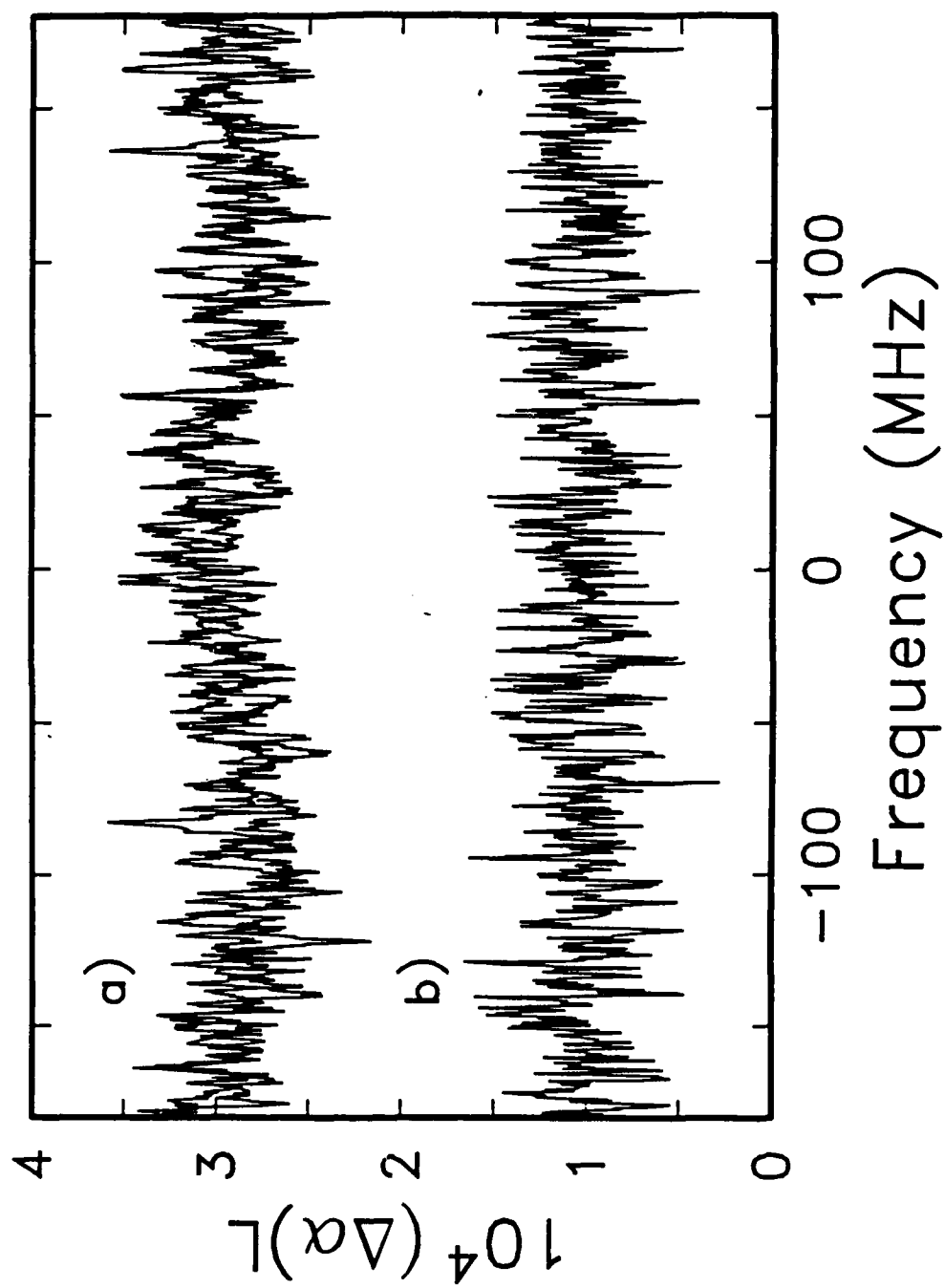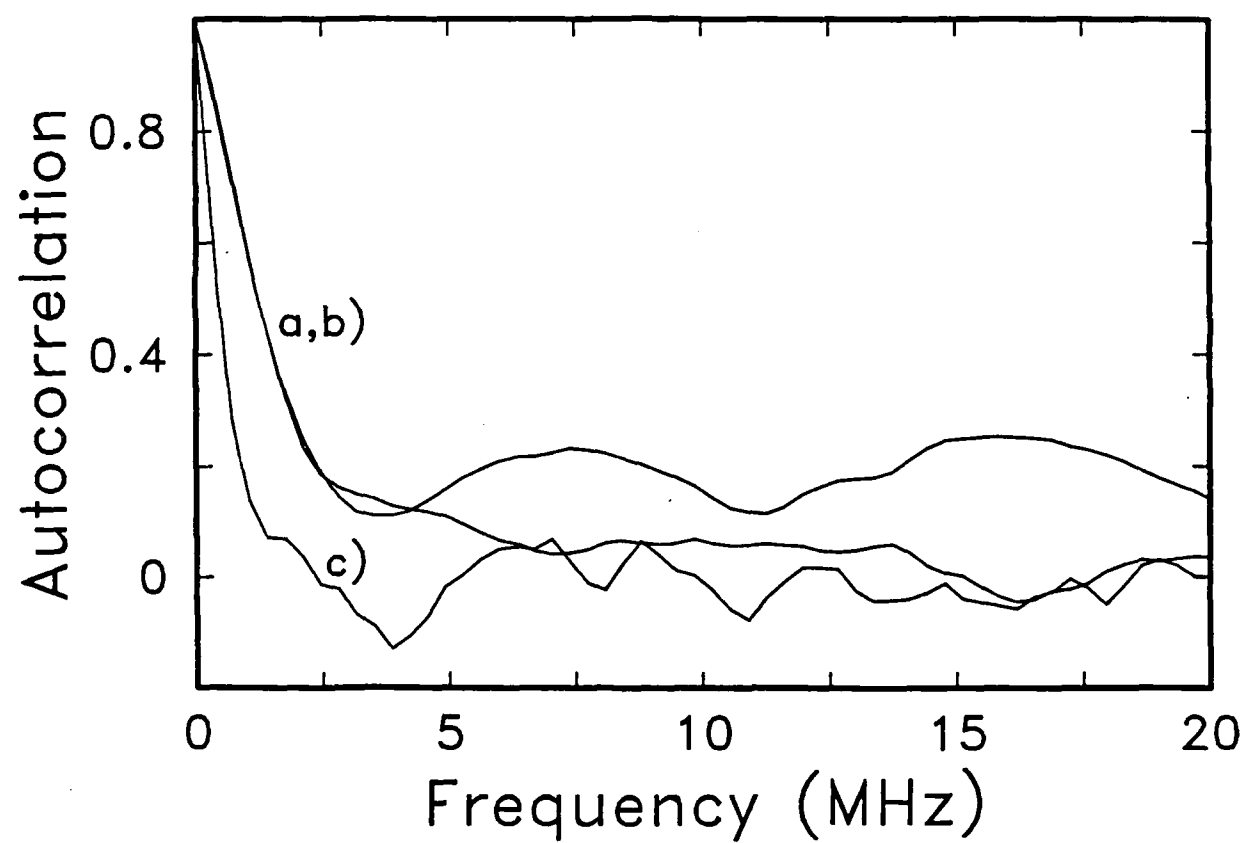
Figure 5

Figure 6

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