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To cite this version:
S. Rand. CONTINUOUS-WAVE PHASE CONJUGATION WITH F-AGGREGATE COLOR CENTERS IN LiF. Journal de Physique Colloques, 1985, 46 (C7), pp.C7-507-C7-510. <10.1051/jphyscol:1985790>. <jpa-00225120>

HAL Id: jpa-00225120
https://hal.archives-ouvertes.fr/jpa-00225120
Submitted on 1 Jan 1985

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CONTINUOUS-WAVE PHASE CONJUGATION WITH F-AGGREGATE COLOR CENTERS IN LiF

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Abstract - We report the first observation of continuous degenerate four-wave mixing (DFWM) with color centers in LiF. Using a new technique which permits subkilohertz resolution, we have also observed the nearly degenerate four-wave mixing (NDFWM) spectrum in the visible spectral region.

I. - INTRODUCTION

In recent years phase conjugation by four wave mixing has been established as an effective nonlinear optical technique for high resolution laser spectroscopy. It has been applied to Doppler-free measurements of level splittings, collision cross-sections and studies of collisional narrowing, dephasing and Zeeman coherence of atoms and molecules. Previously, laser jitter and transit time of resonant atoms passing through the optical beams have limited resolving power however. Also, applications of phase conjugation in gases have been hampered by the practical requirement for operation within narrow resonant frequency ranges, which enhance the nonlinearity of the conjugating medium. Hence there is interest both in new, ultrahigh resolution spectroscopic techniques and for phase conjugators which exhibit efficient four-wave mixing over wide spectral regions.

In this work we describe a new technique for the performance of nearly degenerate four-wave mixing (NDFWM) spectroscopy which is insensitive to laser jitter. We apply it to the first study of continuous-wave DFWM by color centers in a solid and resolve features of the NDFWM spectrum as narrow as 7 Hertz. Defect structures in solids can be created in closely controlled concentrations, with virtually any desired resonance frequency and usually have broad absorption/emission bands due to vibronic interactions with the host lattice. They therefore represent a potentially large class of efficient, broadband conjugators. Their formation is well understood in the alkali halides so that in terms of fabrication they offer an appealing alternative to photorefractive materials.

II - EXPERIMENT

F-aggregate color centers in alkali halides are similar in many respects to more familiar saturable absorbers, such as organic dyes.
The F\textsuperscript{−} center in LiF has a peak absorption at 960 nm, is saturable at 1.06 \( \mu \)m with \( I_s=0.93 \text{ MW/cm}^2 \) and is photostable at room temperature to all but UV radiation\(^2\). The \( \text{F}_2 \) and \( \text{F}_2^+ \) centers have absorptions in the visible spectral region but suffer from photoionization and recombination instabilities respectively. High concentrations of several F-aggregate centers are easily generated in LiF by exposure to ionizing radiation at room temperature, so it is to be expected that four-wave mixing due to saturated absorption, or possibly other mechanisms, can occur in this material over extremely broad spectral regions involving one or more types of defect.

DFWM and NDFWM experiments were carried out at 1.06 \( \mu \)m (pulsed) and 610 nm (cw) in LiF irradiated with 105 MRads of Co\textsuperscript{60} \( \gamma \)-rays to investigate the conjugator potential of F-aggregate centers. Work at 1.06 \( \mu \)m was performed with a novel single-mode Nd:YAG laser containing a LiF passive Q-switch\(^2\). Counter-propagating pump beams were provided simply by reflection of 0.5 mJ incident pulses with a plane mirror beyond the sample, as in Ref. 3. Approximately 20\% of the incident power was split off for use as a probe making an angle of 1.9 degrees with the pump beams. A probe beamsplitter directed the backward-wave through a small aperture and neutral density filters to a photomultiplier. In a sample with an absorption-length product of \( \alpha_L=0.2 \), a cubic dependence of the signal was verified up to 0.25\( \mu \)s, at which point saturation began to limit the mixing, as expected. A maximum reflectivity of less than 1\% was obtained. With \( \alpha_L=0.3 \), signals were 16 times smaller and for \( \alpha_L=0.09 \) no signal was observed. The phase conjugate nature of the signals was tested simply by insertion of a lens aberrator in the probe beam to verify that the signal beam passing back through the probe aperture remained constant, apart from reflective losses at the lens surfaces.

![ ARGON PUMPED DYE LASER

\begin{align*} 
\text{CR 590} & \quad \omega_L \\
\text{WITH ETALONS} & \\
\text{SAMPLE} & \quad \omega_L + \omega_1 \\
\text{DET.} & \quad \text{SIG. OUT} \\
\text{CHOPPER} & \\
\text{DUMP} & \quad \omega_L + \omega_2 \\
\text{A.O. MOD.} & \\
\text{FREQUENCY SYNTH.} & \phi \text{ LOCK} \\
\text{FREQUENCY SYNTH.} & \omega_1 \\
\end{align*}

Figure 1. Experimental configuration for NDFWM using synthesized frequencies for the three input beams.

At 610 nm, continuous four-wave mixing due to F-centers was observed in pure and doped LiF with the apparatus shown in Fig. 1. Most measurements were performed in a sample of 0.4\% Li\textsubscript{2}0:LiF having \( \alpha_0L=1.24 \). Quadratic growth of reflectivity was observed up to an
effective intensity of 0.5 W/cm², above which slow saturation of the signal was evident. Maximum observed reflectivity was 10⁻³. Phase conjugation was readily demonstrated by restoration of the signal beam in the presence of severe probe beam distortion (Fig. 2).

Figure 2. (a) Input beam photographed with a plane mirror at the sample position (b) Aberrated input beam (c) Restored input beam with conjugator and aberrator in place.

The NDFWM spectrum shown in Fig. 3 was obtained by scanning the synthesized probe frequency over a 200 Hertz interval centered at the frequency of the pump beams. The spectrum consists of three peaks separated by 10 Hertz with full widths of 7 Hertz. Neither the splittings nor the widths exhibit any dependence on external magnetic fields or the optical intensity, ruling out involvement of magnetic substructure and the AC Stark effect. No signal was observed for "cross-polarized" pump and probe beams, which in atomic vapors can only arise from Zeeman coherence among magnetically degenerate sublevels.

Figure 3. NDFWM signal as a function of pump-probe detuning, δ.

III. - DISCUSSION
The pulsed DFWM results at 1.06μm are completely consistent with mixing due to saturated absorption of F₂⁻ centers. These results are in essential agreement with those of Ref. 3 and are in the transient regime since the F₂⁻ decay time τ_f = 64 ns² exceeds the excitation
pulsewidth. Higher four-wave mixing efficiency may be expected with longer pulses, larger sample $a_0L$ values and balanced pump beam intensities.

The continuous-wave mixing at 610 nm, on the other hand, occurs at intensities far below the calculated off-resonant saturation intensity $I_s(\nu)=160 \text{ kW/cm}^2$. While the interaction could be due in principle to photorefraction, LiF has a very small electro-optic coefficient and we have observed no beam coupling in 2-wave experiments with phase-sensitive detection, rendering this mechanism unlikely. Also, the sample gratings decay in 45 ms, so that thermal gratings in the lattice which decay in microseconds cannot play a role.

The narrow resonances observed in the NDFWM spectrum support the picture of involvement of a third, metastable level. The linewidths are much narrower than the inverse decay time of $F_2$ centers, which is $(13 \text{ ns})^{-1}$, and correspond to $1/\pi T$, where $T$ is the measured grating decay time of 45 ms, noted above. In Cr:YAlO$_3$ the same spectroscopic method was applied recently to show that relaxation of the ground state spatial hole-burning was entirely accounted for by fluorescent decay of the $^2E$ metastable level, giving a Lorentzian NDFWM spectrum with a linewidth of 9.8 Hertz. In the latter case mixing is due to the first term in the signal intensity, calculated by third order perturbation theory:

$$I_s \propto \frac{\gamma_r^2}{(\gamma_T-\gamma_{f1})^2} \left[ 1 + \frac{\gamma_{sp}-\gamma_{f1}}{\gamma_T-\gamma_{f1}} \gamma_{f1}\gamma_T \right] \frac{1}{(\gamma_T+\gamma_{f1}) \delta^2+\gamma_{f1}^2}$$

In this expression $\delta$ is the probe detuning and the $\gamma_i$ are three-level relaxation rates.

In LiF, there is no similar metastable fluorescent level to explain nonlinear mixing at low intensities. However a long-lived triplet spin state which can account for our observations has been seen by EPR in optically excited $F_2$ centers and studied by Seidel. A three-level model is therefore justified and the results reported here may be explained by saturated absorption, with the multiresonant NDFWM spectrum possibly being related to additional contributions arising from the dispersive terms in the nonlinear polarization.

Acknowledgement: The author wishes to thank Drs. G.C. Valley, R.N. Schwartz, R.A. McFarlane and D.G. Steel for helpful discussions of this work.

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