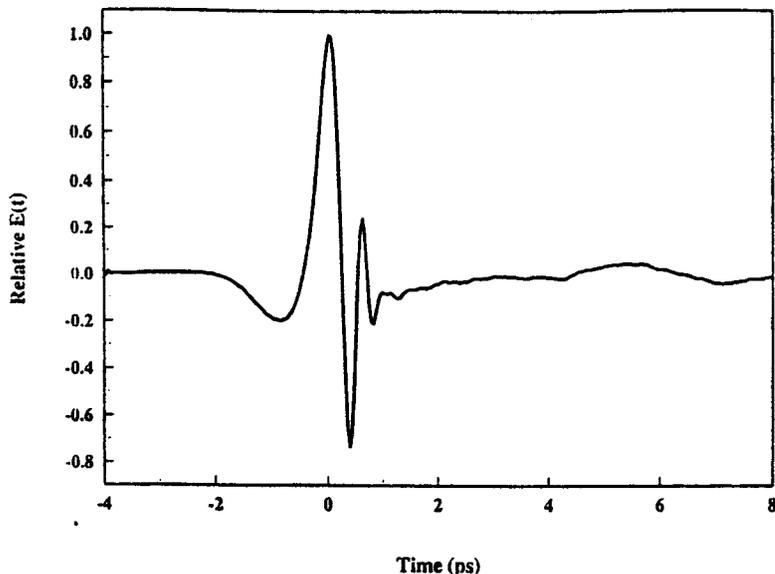


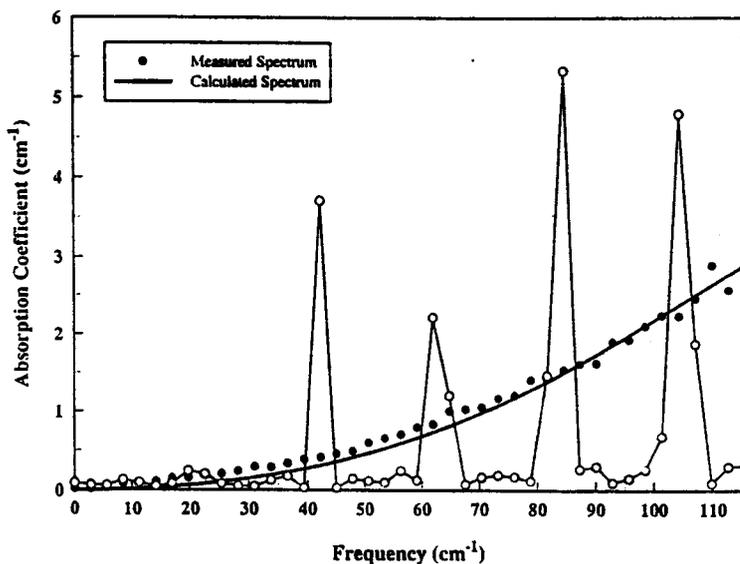
Pulsed terahertz study and spectral analysis of a simple solution: HCl in CCl₄

B. N. Flanders, P. Moore, R. Cheville,* M. Klein, D. Grischkowsky,* N. F. Scherer,
University of Pennsylvania, Department of Chemistry, Philadelphia, Pennsylvania 19104-6323

Recent advances in generation of high intensity radiation in the far-infrared (FIR) spectral region¹ have suggested the possibility of performing a FIR spectral hole burning measurement to determine the homogeneous rotational dephasing time of a dipolar rotor in condensed media. Knowledge of the individual rotational line widths and shifts would be needed for such a study. This paper describes the first steps, in terms of both measured and calculated line shapes, in studying



QWD46 Fig. 1 Measured THz pulse.



QWD46 Fig. 2 Experimental and calculated frequency-dependent absorption coefficient are represented by solid circles and the boldface curve. The gaseous HCl frequency-dependent absorption coefficient is represented by the lighter faced curve and hollow circles.

densed media. The advances in measuring and understanding far-IR relaxation behavior are demonstrated for the simple solute-solvent system HCl in CCl_4 .

The THz time-domain wave forms are measured in transmission mode.² The temporal profile of the THz electric field is shown in Fig. 1. The dilute solution of HCl in CCl_4 is prepared by bubbling HCl(g) into liquid CCl_4 . The solution is transferred to a 1-cm sample cell with silicon windows for measurement. The HCl/ CCl_4 - pure CCl_4 frequency-dependent absorption coefficient is shown in Fig. 2.

The spectral theory³ used here determines the FIR dipolar absorption spectrum to be a superposition of spectrally broadened and

the anisotropic potential time auto-correlation function (APTAF). Although on average the solution is isotropic, the anisotropic intermolecular potential is important because anisotropy in the orientational fluctuations of the HCl molecules is determined by the interaction between the HCl dipole and the surrounding solvent molecules. These solute-bath interactions are responsible for the rotational line broadening and shifting observed in the experimental data.

The APTAF may be determined by molecular dynamics simulation of the system and bath and experimentally determined anisotropic potential parameters from gas phase microwave and radio frequency spectra,⁴ although for the HCl/ CCl_4 case a stochastic model is

and has been shown to approximate the time scale of the decay and shape of the simulated correlation function reasonably well.⁶

The calculated fit normalized to the experimental data is shown in Fig. 2. A best fit was obtained when the interaction strength parameter $\lambda^2 = 165$ and the APTAF decay time $\tau = 0.10$ (in reduced units where $\lambda^2_{\text{reduced}} = \lambda^2_{\text{fit}} / (\text{Bhc})^2$ and $\tau_{\text{reduced}} = (2\pi\text{Bc})\tau_{\text{fit}}$). These results are qualitatively consistent with FT-IR spectra and simulations of HCl in Ar and SF_6 . While CCl_4 is most similar in mass to SF_6 , the polarizability of 11.2 is almost twice the value of SF_6 and seven times the value of Ar. A larger solvent polarizability means that the dipolar solute induces a proportionally larger dipole moment on the neighboring solvent molecules. Hence the dipole-induced dipole interaction between HCl and CCl_4 is stronger than between HCl and SF_6 and much stronger than between HCl and Ar. This trend suggests that the stronger the dipole-induced dipole interaction in the solution, the greater the degree of line broadening and shifting in the corresponding absorption coefficient spectrum. From these results we conclude that THz hole burning studies should be carried out in low polarizability (even atomic) solvents at low temperature.

**Oklahoma State University, School of Electrical and Computer Engineering, Stillwater, Oklahoma 74078*

1. D. You, R. R. Jones, P. H. Bucksbaum, D. R. Dykaar, *Opt. Lett.* **18**, 290 (1993).
2. N. Katzenellenbogen, D. Grischkowsky, *Appl. Phys. Lett.* **58**, 222 (1991); B. N. Flanders, R. A. Cheville, D. Grischkowsky, N. F. Scherer, *J. Phys. Chem.* **100**, 11824 (1996).
3. S. Mukamel, *Chem. Phys.* **37**, 33 (1978).
4. S. L. Holmgren, M. Waldman, W. Klemperer, *J. Chem. Phys.* **67**, 4414 (1977); *ibid.*, **69**, 1661 (1978).
5. A. Medina, S. Velasco, A. C. Hernández, *Phys. Rev A* **44**, 3023 (1991); *ibid.*, **45**, 5289 (1992).
6. A. Medina, A. C. Hernández, S. Velasco, *J. Chem. Phys.* **100**, 252 (1994).

QWE

2:30 pm-4:00 pm
Room 337

Atomic Interference

John E. Thomas, *Duke University, President*

QWE1 (Invited)

2:30 pm

Atom holography

Fujio Shimizu, *University of Tokyo, Japan*

We generated an ultracold Ne atomic beam in the $1s3$ ($J = 0$) metastable state by optical pumping of the $1s5$ atoms stored in a magneto-optical trap. The atomic beam was then diffracted by a computer-generated binary hologram and the resulting interference pattern was measured. The binary