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TITLE EXPERIMENTAL MOLECULAR SPECTROSCOPY IN SHOCK-COMPRESSED MATERIALS

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EXPERIMENTAL MOLECULAR SPECTROSCOPY IN SHOCK-COMPRESSED MATERIALS*

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Shocked materials present an exciting challenge to the experimental spectroscopist. The severe constraints in time and space imposed by the shock process can also be accompanied by chemical complexity. Many standard spectroscopic techniques have recently been adapted with varying degrees of success. Those discussed here are uv/visible absorption, emission, and fluorescence, infrared absorption, and spontaneous and coherent Raman scattering. The importance of the connection to both theory and static high-pressure methods is emphasized.

1. INTRODUCTION

Advances in the understanding of shock/ material interactions have paralleled the advances in the experimental techniques available to probe the sample. The measurement of bulk properties such as density, temperature, mechanical deformations, and hydrodynamic effects have provided useful characterizations of some materials, but there remain difficulties in the performance and interpretation of such experiments in molecular materials, especially energetic ones. The purpose of this paper is to review the various spectroscopic techniques that have been or could be used to measure molecular-level properties of shock-compressed molecular materials. In particular, we are interested in the use of such techniques to identify the molecular physical and chemical processes that occur upon shock compression and how these microscopic processes couple to hydrodynamics and energy transport. In this context several necessary capabilities of the experimental probes can be discerned. (1) they need to be able to distinguish between chemical species; (2) they should be able to provide structural information; (3) they should have enough temporal and spatial resolution to measure the desired kinetic behavior, and (4) they should

operate in real time.

There are many experimental difficulties to overcome to adapt optical techniques to the study of shock-compressed materials. For many materials, strong shock waves are believed to be less than 1 μm in thickness and have velocities of the order of 5 km/s. Therefore, if we desire to make spectroscopic measurements in the shock wave, the technique selected needs spatial and temporal resolution of better than 0.2 μm and 10 ps, respectively. However, these requirements are considerably relaxed to study the region immediately behind the shock wave. Under shock-compression many materials are opaque or become opaque. Consequently, the use of these optical techniques may be limited to phenomenological studies in a few select materials. Another limitation is the refractive index change through the shock that results in bending of optical paths. Therefore, we may be forced to use optical arrangements that are not necessarily optimum for each technique, and may not be able to use some techniques at all. We also desire that the optical probe be non-intrusive. The measurements may be distorted by probe-induced effects such as photochemistry. Finally, in inhomogeneous samples, measurements will often be averaged over the inhomogeneities.

and consequently will not reflect the details of
the microstructure. This effect may also occur
in samples thought to be homogeneous.[1] It
should now be obvious that no one optical
technique has all of the necessary spectroscopic
qualities or could overcome all of the above
experimental difficulties. Therefore, the
spectroscopic study of these systems will
necessarily be multifaceted, using a combination
of several techniques in appropriately chosen
select materials.

2. TECHNIQUES

2.1. Ultraviolet/Visible Methods

a. UV/visible Emission Spectroscopy of
condensed-phase, shock-compressed materials has
been reported by many authors. These
measurements have been used to determine
temperatures[2-4] and to identify spectral
features.[2-16] Other investigations of emission
during shock-compression have been reviewed by
Davison and Graham.[17] The calculation of
temperature by using either brightness or multi-
color methods usually requires additional
knowledge or assumptions about the emissivity
and optical depth of the radiating source.
Failure to recognize this can result in an
incomplete interpretation of the observations
For example, a 6000 F black body source viewed
through a 5000 K black body will not be observed
since the radiation is absorbed and re-emitted
at the temperature of the intervening layer
This effect has been observed during the
compression of ionic crystals[2] Also, what do
temperature measurements in compressed powders
mean? Does the emission originate because of
friction between microcrystals at the voids or
compression of gas in the voids, or is it
indicative of a bulk temperature within the
microcrystals?

Some spectral features differing from gray
body emission have been detected. Among these
are the luminescence in shock-compressed

quartz,[2,5,8] molecular spectral features in
shock-compressed RDX and PETN,[3,6,7] shock-
compressed benzene,[9,10,15] excimer-laser-
initiated lead styphnate and mercury
fulminate,[11] detonating HNS, HMX, and PETN,[10,15]
and shock-compressed granular HNS and PBX
9404.[13,16] The emitting species that have been
identified include C_2, CN, CH, NO_2
(tentatively),[10] and many atoms such as Na.
While in some cases the emission is admittedly
from gas phase products and can be understood in
terms of gas phase molecular parameters, in
other cases the emission arises from within the
condensed sample.[9,12,14] A pressing question is
how the emission features shift and broaden with
pressure and chemical composition. In addition,
the origin of molecular emission in granular
explosives may be ambiguous as discussed above.
Other serious questions are what is the role of
spalled material, and what high-temperature,
gas-phase reactions influence the result?

b. UV/visible Absorption has been used to
study index of refraction changes,[4,17] and the
shifts of absorption bands with pressure.[18-21]
Many improvements to the UV-visible absorption
technique have been implemented to overcome
experimental ambiguities. The latest
refinements, one with a flashlamp in a
projectile, and one with a reflecting surface
removed from shock effects, implemented by
Duvall, et. al. are depicted in Fig. 1. The
measured absorption band shifts are used to
infer changes in the molecular energy levels
however, these inferences are complicated,
especially in larger molecules, by the
complexity of the pressure induced shifts in the
molecular states. Recently, dilution studies,
and comparisons to static high pressure
absorption experiments have been used to
tackle some of these ambiguities. In addition,
some progress has been made in modeling the
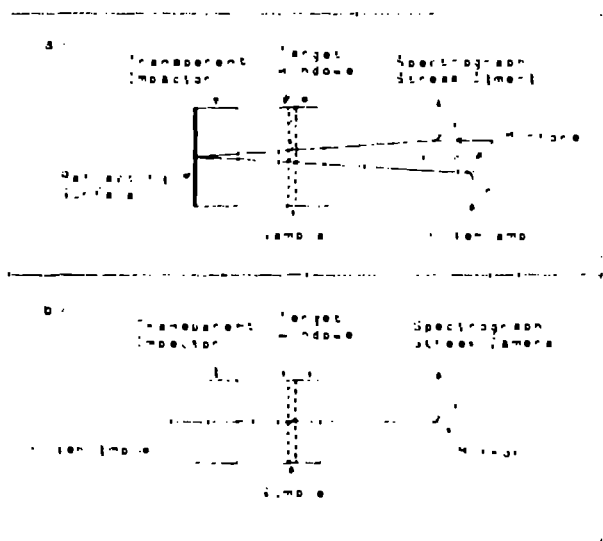density dependence of electronic absorption
bands.[21]

FIGURE 1
UV/visible absorption apparatus with a)
reflector or b) flashlamp in the projectile.[23]

FIGURE 2
Fluorescence spectroscopy apparatus with laser
shock production.[32]

c. Fluorescence is frequently used as a
diagnostic for state-to-state chemical dynamics
studies, measurements of intramolecular
relaxation processes, and determinations of
collisionally driven intermolecular relaxation
rates. Current techniques use lasers as sources
to selectively populate a small subset of
excited states. Subsequent emission from the
manifold of states populated by collisional or
unimolecular processes is then monitored by a
variety of fast detection methods. The
character of the observed emission is determined
by the energy transfer rates, the radiative
lifetimes of the populated states, and the time
resolution of the detection system. Recently
this method (see Fig. 2) has been used to
measure the viscosity and temperature of shock-
loaded molecular systems,[30,32] to study the
shock behavior of a simple diffusion-controlled
chemical reaction,[13] and to measure wavelength
shifts of R line emission in shock-compressed
ruby.[35,35] The problem of collisional quenching
of excited states can be avoided by use of
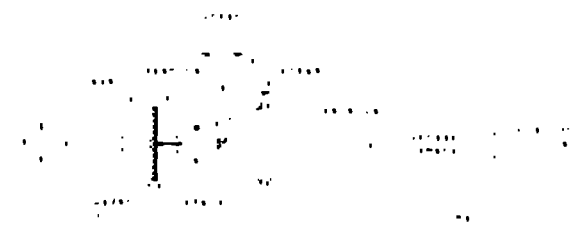molecules well suited to such environments, such

as organic dyes, and, in the region of lower
shock pressures, spectral broadening can be
small.[30-33] However, at higher pressures, even
if pressure- and temperature-induced broadening
is not a problem in specific cases, the dye
molecules themselves may decompose. The key to
the ruby experiments[34,35] is that the absorption
is broad, but the excited Cr^{3+} ion quickly
relaxes into a state whose fluorescence remains
narrow to high pressures and temperatures.
Other transition metal ions (or complexes) may
have the same attributes.

An alternative to normal fluorescence is the
use of double resonance techniques.[36] An
initial pulse could be used to excite a desired
electronic or vibrational transition. A second
laser pulse, temporally delayed with respect to
the first, could then excite the molecule to a
higher energy level. The emission of the higher
levels would then be monitored versus the delay
time between pulses to determine the relaxation
time of the intermediate level.

2.2 Infrared Methods

a. Infrared Absorption techniques were
complicated until recently by the lack of fast
detectors and/or fast pulsed sources. Now,
however, pulse fast nanosecond detectors are
available. To be used effectively, however, a
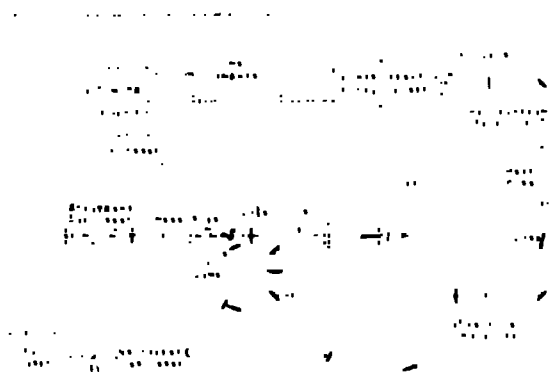very high brightness source is required to

FIGURE 3
Time-resolved infrared spectral photography
(TRISP) apparatus for shocked materials.[38]

FIGURE 4
Pictoral representations of Raman and resonance
Raman processes.

overcome the brightness of the shock-compressed
sample. In addition, for detection of molecular
species, what is needed is an IR detector array,
similar to the intensified photodiode arrays or
vidicons available for work in the visible.
These, unfortunately, are not yet available. A
usable alternative is the time-resolved infrared
spectral photography (TRISP) system devised by
Sorokin and coworkers [37] This system
circumvents both of the above problems by
employing stimulated electronic Raman scattering
in alkali metal vapors to down-convert a
broadband visible laser pulse into the infrared
This broadband infrared pulse is passed through
the sample, recording the absorptions present
there, and is then up-converted back to the
visible, retaining the recorded absorptions,
using a four-wave mixing process also in alkali
metal vapors. The up-converted broadband
visible pulse is then detected using a vidicon
or intensified diode array

TRISP has recently been used in shock-
compressed samples and as a diagnostic to look
for species in the gas-phase plume directly
above detonating small pellets of high
explosives [18] The experimental apparatus for
TRISP measurements in shock-compressed samples
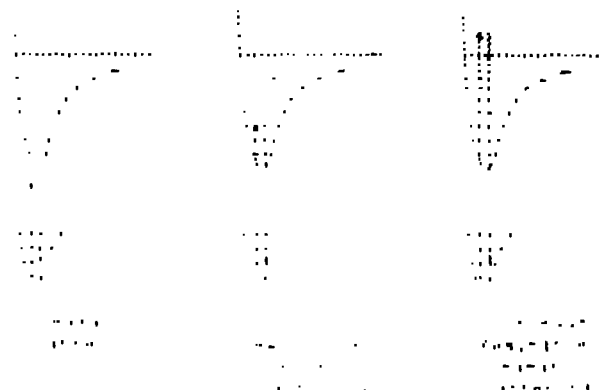is shown in Fig 3 A difficulty with the use

of this technique as a real-time optical
diagnostic in shock-compressed samples is the
timing constraint placed on the method by
necessarily very thin samples (timing jitter
must be much less than 1 ns for use of the 1 ns
IR pulse to measure absorption in a 5-μm-thick
sample). An alternative is to use dilution
methods to avoid saturation of the absorption,
but, as in the case of uv/visible absorption,[25]
dilution may eliminate the concerted-type
chemical reactions that we wish to observe.

2.3. Raman Methods

a. Spontaneous Raman Spectroscopy in shocked
materials was first performed in detonating
crystalline RDX.[6] Subsequently, the technique
has been used to measure the temperature of
detonating[39] and shock-compressed[40] nitromethane
and to observe shock modification of vibrational
bands in detonating PETN,[41-42] HMX,[43] and
RDX,[41-42] shock-compressed water,[45] and shock-
compressed TATB.[49]

Raman spectroscopy is made possible by the
inelastic scattering of a photon by molecules in
the probed sample (see Fig 4). The difference
in frequency between the incident and scattered
photons is equal to the frequency of a Raman
active molecular transition Raman scattering
cross-sections are many orders of magnitude

smaller than those for emission and absorption;
hence, difficulties could be encountered in
materials shock-compressed to very high
temperatures. The cross-sections can be
increased by using UV pump lasers rather than
ones in the visible (the cross-section increases
as ν^4); however, care must be taken to avoid
interferences from fluorescing or
photochemically produced species.

    b. Resonance Raman. When the exciting laser
frequency is in resonance with an actual
molecular transition, the scattering cross-
section for that species can dramatically
increase.[47] In an isolated molecule, the
resonance Raman effect can be due to (1)
interaction of the excitation with a discrete
transition in the molecule, or (2) interaction
of the excitation with a dissociative continuum
in the molecule (see Fig. 4).

    As the excitation energy approaches a
molecular resonance, the distinction between
Raman scattering and absorption and reemission
becomes blurred. What is important to the
distinction between these two processes is the
degree of loss of phase information in the
experiment. In the condensed phase, especially
when shocked to high pressures and temperatures,
it is likely that most molecules will undergo
phase interruption due to collisions within
their normal excited state lifetime implying
that discrete resonance Raman would be like
fluorescence and can be said to have the same
advantages and disadvantages.

    When dissociative transitions are probed,
the gain in cross-section due to the resonance
condition may be more than offset by the small
fraction of excited molecules that re-emit
before dissociating.[4?] Although, in gas-phase
studies, resonance Raman techniques and analogs
(dissociative emissions) have recently undergone
dramatic growth because they provide unique
information about the dissociative potential
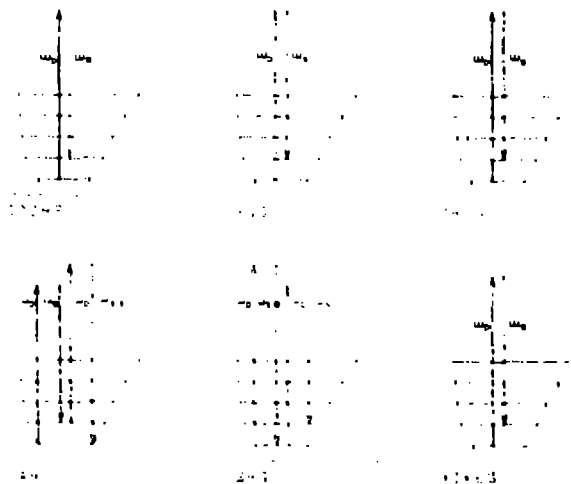energy surface,[48,49] in the study of shock

compressed,condensed-phase systems their use
will likely be problematic.

    Large increases in Raman scattering cross-
sections can also be produced when the exciting
laser is just near resonance with an electronic
transition in the molecule. This near-resonance
enhancement in the Raman cross-section may be
much larger than the normal Raman ν^4 frequency
dependence, and may enable detection of some
species that have the correct spectroscopic
attributes.

    c. Coherent Raman Spectroscopy. Several
coherent Raman techniques have been developed in
recent years (Fig. 5). These techniques use
large optical frequency electric fields to non-
linearly drive the inelastic scattering process,
leading to large scattered signals that are
beam-like in nature. The consequent demands on
the signal collection and detection systems are
therefore significantly reduced from those
required by spontaneous Raman methods.

    Stimulated Raman was the first coherent
Raman technique to be used in a shock compressed
sample.[50] Stimulated Raman scattering[51] occurs
when the incident laser intensity in a Raman-

active material exceeds a threshold level and generates a strong, directional (forwards and backwards), beam at the Stokes frequency. The backwards directed beam was conveniently used in shock experiments to minimize the experimental complexity.[50] This technique does have some disadvantages: (1) the incident laser powers are large; (2) not all molecules have the required large Raman cross-section and narrow Raman linewidth; (3) only the lowest threshold transition in the condensed sample produces a signal; and (4) spatial resolution is limited to the confocal parameter of the focusing lens.

Inverse Raman or stimulated Raman loss spectroscopy (SRLS), and its analog stimulated Raman gain spectroscopy (SRGS),[52] have been suggested[53,54] as diagnostic techniques for shock-compressed materials. Signals can be obtained at power levels considerably below those required for stimulated Raman scattering because the process is driven by the presence of both the incident and Stokes laser fields (see Fig. 5). In the process, some fraction of pump photons is converted into Stokes photons. The difference in the two techniques lies in which beam is detected. In the case of SRLS, the loss of photons at the pump frequency is detected. In SRGS, the gain of photons at the Stokes frequency is detected. The lack of accurate information on vibrational frequencies at elevated pressures and temperatures, and the desire to detect more than one species, dictates the use of a broadband laser for one of the two incident pulses. The limiting detectivity is determined by the spectral noise in the broadband laser unless signal averaging or double beam techniques are used.

Coherent anti-Stokes and coherent Stokes Raman scattering (CARS and CSRS, respectively)[51] (see Fig. 5) occur as four-wave parametric processes in which three waves, two at a pump frequency and one at either the Stokes (CARS) or anti-Stokes (CSRS) frequency are mixed in a
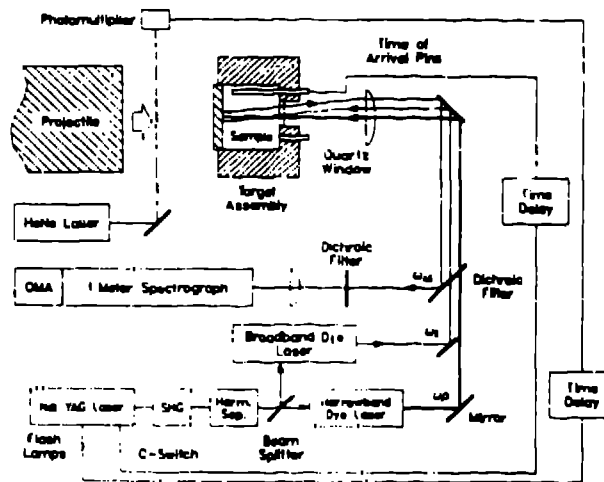

FIGURE 6
Coherent anti-Stokes Raman (CARS) apparatus for shocked materials.[55]

sample to produce a coherent beam at the anti-Stokes (CARS) or Stokes (CSRS) frequency, respectively. This mixing is greatly enhanced if the frequency difference between the pump and the Stokes (or anti-Stokes) lasers coincides with a Raman active mode of the sample. Like SRGS and SRLS, CARS and CSRS can be produced at incident power levels considerably below those required for stimulated Raman scattering. However, in CARS and CSRS, phase-matching is required, and possible geometric arrangements of the beams are limited. Figure 6 is a diagram of an experiment used to measure CARS spectra in shock-compressed liquids.[55-58] The noise in the broadband, Stokes, dye laser spectral profile also causes problems with the CARS measurements, but does not prevent detection of the spectrum, as it might in SRGS or SRLS techniques. To interpret the data, the spectral profile of the broadband dye-laser pulse is recorded at the same time as the CARS spectrum using another spectrometer/photodiode array.[57] Measurements that have been made to date using the CARS technique include the pressure and temperature shifts of the ring-stretching mode of benzene to 14.5 GPa,[55-58] the CN stretching mode of

nitromethane to 9.0 GPa,[56,59] the fluid N_2
vibration fundamental and hot-bands to 19 GPa
single shock and 41 GPa double shock,[57,58] and
the fluid O_2 vibration to 9 GPa.[58] These
results have shown that CARS measurements can
provide very accurate molecular level
information at pressures and temperatures of
interest to both the shock-wave and equation-of-
state communities. For example, the data have
been used to test theoretical estimates of
vibrational frequency shifts and bond lengths in
these dense molecular fluids.[60] Through such
theoretical modeling, it should be possible to
characterize molecular potential functions at
very high densities and temperatures.

In CARS experiments, the temporal resolution
is determined by the laser pulse duration.
However, since the beam crossing angle is
determined (at from 1-3 degrees) by phase-
matching requirements, spatial resolution is
quite poor, being of the order of the confocal
parameter of the focusing optics. One advantage
of CARS and CSRS experiments is the large
signals produced, allowing discrimination
against background fluorescence and emission.
However, CARS and CSRS produce signals even when
there is no vibrational resonance, which is a
problem when attempting to detect small
concentrations of species. The resonant and
non-resonant parts interfere to produce
asymmetric line shapes, and ultimately, the
resonant contribution will be lost in the noise
(at the ppt level). However, the asymmetric
lineshape has so far been used to advantage when
fitting spectra to determine peak positions and
linewidths.[59] Still unexplored is the
possibility of using nearby electronic
transitions to resonantly or near-resonantly
enhance the CARS intensities (as was discussed
above for spontaneous Raman).[61] However,
considerable theoretical effort into the effects
of broadening electronic and vibrational
transitions on CARS lineshapes and intensities

needs to be undertaken before resonantly
enhanced CARS can become a truly useful
experimental tool.

Raman-induced, Kerr-effect spectroscopy
(RIKES)[52] has been suggested[53] as a diagnostic
technique for performing measurements in shock-
compressed systems. RIKES uses a circularly
polarized pump beam and a highly linearly
polarized broadband Stokes beam, and requires no
phase-matching. At conditions of Raman
resonance, the interaction of the two beams
results in slight elliptical polarization of the
Stokes beam at the Raman resonant frequencies.
This elliptical component then passes through a
crossed polarizer, and is detected by a
spectrometer/intensified diode array. This
arrangement ideally would give large signals
against a small background, making it a good
choice for the detection of species with small
concentrations. In shock applications, however,
difficulties due to birefringence in the
strained shock-compressed sample, or
polarization rotation on reflection from a
shocked mirror could result in detection of many
incident probe frequencies irrespective of a
Raman resonance, and would give a large unwanted
background.[62]

3. SUMMARY

We believe that the use of optical probes in
shock-compressed materials will continue to
spread and increase. The techniques most
revealing of molecular properties, such as Raman
scattering, will give specific information
regarding shock-induced structure changes,
chemical reactions, etc. This information, when
used in conjunction with the very important
recent[60,63,64] and future theoretical efforts,
as well as established bulk property
experiments, will provide insight into the
detailed microscopic processes through and
immediately behind the shock front.
Complementary high pressure/high temperature

static measurements will be used to test diagnostic tools, determine structure and energy transfer rates at shock-compression conditions, and, hopefully, help to distinguish between shock and static environments.

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