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Optical pulses in tritiated solid hydrogen

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We report light-emission data from a solid deuterium-tritium sample between 2.4 and 4 K. We have measured the relative intensity as a function of temperature both from a steady-state background and pulsed flashes which we stimulate with a small heater. The pulse of light is believed to be related to heat spikes previously observed in tritiated solid hydrogen and allows a study of the dynamics of this process without coupling to the apparatus. The pulse of light does not have a sharp cutoff with temperature, but decreases in intensity and increases in duration with increasing temperature such that the area under the pulse is roughly constant between 2.4 and 4 K. We are able to confine most of the light to wavelengths longer than 780 nm.

I. INTRODUCTION

For some time we[1,2] and others[3] have studied the accumulation of unpaired atoms in condensed tritiated mixtures of hydrogen isotopes. These experiments have shown that the β decay of the tritium breaks molecular bonds and produces unpaired atoms which recombine slowly in solid hydrogen. Below 10 K the population of these atoms increases rapidly and can reach levels higher than 1000 ppm. Webeler[4] has observed unstable recombination of these atoms using thermometry, and we have studied the same phenomena using electron-spin-resonance techniques to measure the atom population before and after the recombination spikes.[1] This process has been modeled by Rosen[5] and Zeleznik[6] as oscillations between the stored-atom populations and the enthalpy of the solid. A difficulty in characterizing the thermal behavior of the heat pulse is deconvoluting the effects of the apparatus. In Zeleznik's model, the interesting experimental quantity is the sample temperature. This can be written

$$\frac{dT}{dt} = \frac{-(T-T_0)}{\tau} + \alpha K m^2 ,$$

where T_0 is the temperature to which the sample would cool without a heat load, τ is the characteristic cooling time of the apparatus and sample, m is the concentration of unpaired atoms, $2K$ is the recombination rate of the atoms, and $\alpha = q/C_v$, where q is the recombination energy and C_v is the heat capacity of the system. Since the heat capacity of the system may be dominated by the apparatus rather than the sample, the measured temperature change is only an indirect indication of the behavior of the sample. In this paper we will describe the emission of pulses of light from solid deuterium-tritium (DT) mixtures which we believe are associated with the previously studied recombination phenomena. The time scale of these pulses is much shorter than the thermal response time of the apparatus and results in much smaller temperature changes than those seen in previous experiments, suggesting that the bulk of the energy escapes as light.

II. EXPERIMENTAL RESULTS

The experimental sample cell was a 15-mm-diam spherical sapphire shell with a 1.2-mm-thick wall. A 0.25-mm-diam fill tube was glued into the top of the ball and a chip of germanium of the type used in commercial low-temperature thermometers was glued to the ball near the fill tube and used as a thermometer. This assembly was hung in the center of a 200-mm-diam chamber containing helium exchange gas to cool the sapphire cell. Several calibrated germanium resistance thermometers were mounted on the exchange gas chamber and were used to calibrate the germanium chip on the sapphire cell before loading it with DT. In all cases the hydrogen sample consists of a 50-50 mixture of deuterium and tritium (25% T_2, 25% D_2, 50% DT). The exchange gas chamber is cooled by a variable-pressure helium-gas cell which is in contact with the liquid-helium bath of the Dewar. With this arrangement the temperature of the exchange gas chamber can easily be varied from 2.1 to 30 K. The cell is partially filled with about 3.8×10^{-3} moles or about 0.075 cm^3 of liquid DT at 20 K before cooling to lower temperatures. The decay heat produced by the tritium heats the ball by about 0.3 K when the exchange gas chamber is cooled to its minimum temperature of 2.1 K. Optical windows in the exchange gas chamber, the liquid-nitrogen shield of the Dewar, and the vacuum jacket of the Dewar allow direct observation of any light emitted by the sample. The effective bandwidth of the windows limits us to an optical domain between about 230 and 2500 nm.

Our studies of the light emission began when we noted that samples cooled below 10 K give off a red glow visible to the naked eye. Figure 1 shows the relative intensity of the sample as a function of temperature. These measurements were made by imaging the ball onto a Photometrics CCD camera with a bandwidth from 450 to 900 nm and reading out the intensity of individual pixels. The rapid increase in intensity below 10 K is similar to the increase in the electron paramagnetic resonance (EPR) signal from DT at these temperatures,[2] and suggests that the accumulation of unpaired atoms plays a role in the generation of the background steady-state light.

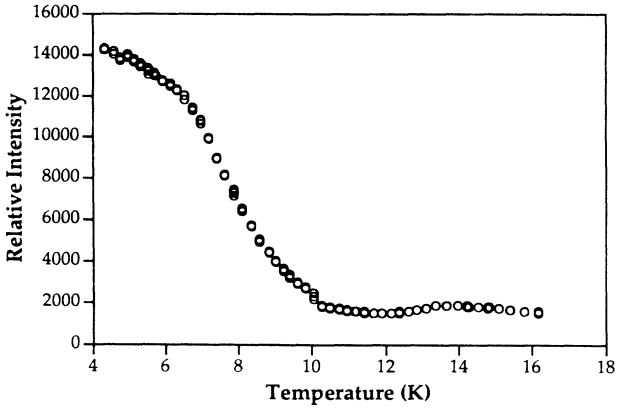

FIG. 1. Relative intensity vs temperature for the steady-state glow of the DT sample.

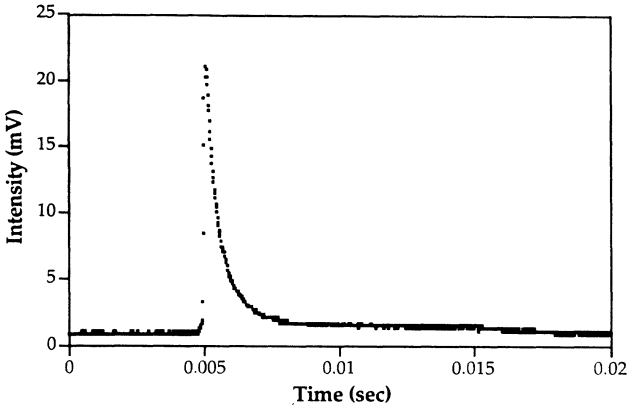

FIG. 2. Intensity of light vs time during a flash at 2.4 K.

The spectrum of the steady-state emission was investigated in the wavelength region of 350–750 nm using a $\frac{1}{4}$-meter polychrometer equipped with a 1200-line grating and a 1024-channel microchannel-plate-intensified scanning-diode array. The only feature observed consisted of a moderate, 10-nm full width at half maximum (FWHM) peak at ~694 nm (undoubtedly due to the ruby R line from a chromium impurity in the sapphire sphere) embedded in a featureless rapidly increasing near-infrared emission, terminated by the long-wavelength cutoff of the detector. No emission was detected in the region from 350 nm to the beginning of the 694-nm ruby peak.

In order to initiate recombination spikes, a 50-Ω heater was mounted in the exchange gas about 2 cm below the cell. A 12-V pulse applied to this heater reliably generates a light pulse as long as the previous light pulse took place at least 10 min earlier. The delay between triggering the heater and the appearance of the light pulse ranged from about 200–400 ms. In no case have we observed spontaneous-recombination spikes in this apparatus such as those described by Webeler. We suppose that temperatures lower than 2 K are required to generate this instability. The light pulses were detected by imaging the ball onto a Newport model 882 silicon photodiode with a 50-mm quartz lens. The response of the photodiode roughly covers the bandwidth from 400 to 1100 nm. The photodiode was biased and amplified with a PAR model 113 preamp and stored in a Tektronic 2430 digital oscilloscope before the data were downloaded into a personal computer for analysis. Data were gathered by manually switching on a power supply to the trigger heater at a fixed time interval of 15 min. Before the first flash was triggered, the DT sample formed a thin layer of ice at the bottom of the sample cell. During the flash the sample appeared to move within the cell as though the sample were exploding. After the first several flashes the sample redistributed itself in a thin apparently uniform layer around the sample cell. The calculated thickness of this layer was 150 μm. Figure 2 shows a typical flash measured at a cell temperature of 2.4 K. The spectral composition of this light has been crudely characterized by measuring spikes with a series of filters in front of the photodiode. We find that long-wavelength-pass filters with cutoff frequencies in the visible or near ultraviolet do not attenuate the photodiode response. A long-pass filter with a cutoff at about 710 nm shows less than 10% attenuation, whereas a long-pass filter at 780 nm cuts off about half the light. At this wavelength our detector sensitivity is falling off, so we conclude that the optical pulse is in the infrared with more than half the light at wavelengths longer than 780 nm.

Survey experiments were performed using the diode-array spectrometer, to characterize the recombination-spike emission in the visible and near-infrared (NIR) spectral region. While there was some uncertainty in synchronizing the time aperture of the diode array with the flash, a cleanly captured event showed enhanced emission at the ruby peak compared to the rest of the NIR feature. Immediately following the flash, the steady-state ruby-peak emission was unchanged from that prior to the flash but the NIR feature was reduced about 90% in intensity. Coupled with the results from the long-pass-filter experiments, we confirm that the majority of the emission in the combined-wavelength region is longer than 780 nm and that the NIR feature seen by the diode array is a low-intensity high-frequency tail of a larger-intensity pulse increasing to longer wavelength. Very little of the emission is due to the 694-nm ruby emission peak.

An optical pulse leaves a characteristic thermal signature lasting much longer than the light output. Figure 3 is a plot of the response of the cell thermometer following a recombination spike. The temperature of the sapphire cell drops by about 20 mK and recovers to its previous value exponentially with a time constant of about 560 sec. Just after a flash, the steady-state background light goes away and returns with about the same time constant as the temperature. We believe the cell cools as energy produced by decay of tritium is used to replenish the reservoir of unpaired atoms rather than heating the cell. The thermal response of the cell allows one to make the following simple calculation of the energy involved in a recombination spike. The 0.075-cm^3 fill of DT produces 3.7 mW of decay heat which warms the cell by 0.3 K above the temperature of the exchange gas chamber when it is cooled to 2.1 K. After a recombination spike we can model the change

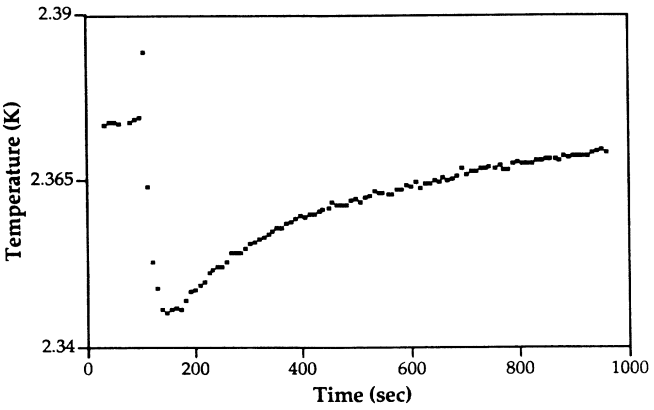

FIG. 3. Temperature of the sample-cell thermometer vs time after a flash.

in the power heating the ball as

$$\delta P(t) = \frac{(0.02K)(0.0037\ \mathrm{W})}{0.3K} \exp(-t/560\ \mathrm{sec}) .$$

Integrating this power over time gives an energy of 0.14 J or 1.8 J/cm^3. At 4.5 eV per atom pair this energy is equivalent to 5.4×10^{18} unpaired atoms/cm^3 or 180 ppm of unpaired atoms in the solid. This value is roughly an order of magnitude lower than EPR measurements of unpaired atom concentrations. The energy collected by the photodiode from the flash is estimated to be 3×10^{-7} J. This gives a total energy radiated by the sample within the bandwidth of the detector of 3×10^{-4} J. This is far less than the 0.14 J calculated above and suggests that much more light may be radiated at still longer wavelengths.

As the cell temperature is increased we find that the amplitude of the optical pulse decreases and its duration increases. Figure 4 is a plot of these parameters as a function of temperature. In all cases the interval between pulses was 15 min. As the cell is warmed the optical pulse is smeared out in time with its duration increasing by almost a factor of 100, but the area under the flash curve remains roughly constant. There is no sharp cutoff in the ability to trigger a flash as the temperature is raised but it becomes increasingly difficult to detect the pulse and trigger the scope.

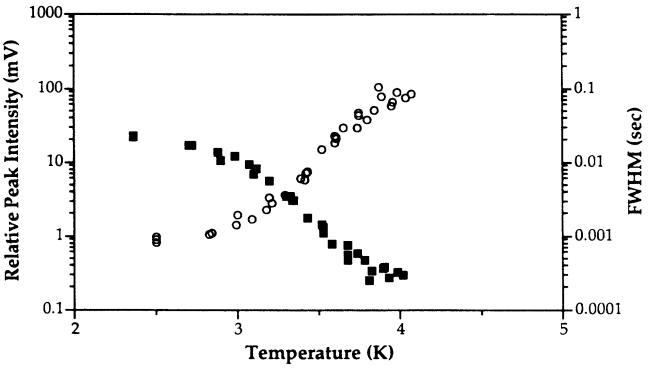

FIG. 4. FWHM (plotted as open circles) and relative peak intensity (plotted as squares) of the optical pulse vs temperature.

The connection between recombination heat spikes in the electron-spin-resonance (ESR) experiment and cold-light pulses from the present experiment is compelling but tenuous. The amount of pulsed light detected in the small-wavelength region from ~710–1100 nm represents just 0.2% of the calculated recombination energy available and may be due to a different phenomenon entirely. Or, as mentioned previously, the remaining energy may be present at longer wavelengths.

The infrared (IR) absorption spectrum of solid tritiated hydrogens consists of several features, described either as radiation or nonradiation induced. Nonradioactive solid hydrogens exhibit well-behaved collision-induced fundamental vibration-rotation spectra. At temperatures below about 10 K, tritiated hydrogens exhibit time-dependent radiation-induced features. One set of these features appear near the fundamental and are Stark shifted to a frequency slightly below the band origin. These features are attributed to either positive-ion or trapped-electron perturbations of the host molecules in the lattice, and appear in a wavelength region beyond 2.5 μm. Another set of strong radiation-induced absorption features, not related to the collision-induced IR spectrum occurs at shorter wavelengths (1.2–2.5 μm) (Ref. 7). This absorption is attributed to electron transitions localized in a bubblelike structure, and is most likely unrelated to the transitions in the host molecule. The light flashes observed in the present experiment may be associated with these trapped electrons. The intense absorption strength of these features may lend a clue as to the dichotomy of heat vs light spikes in the ESR and present experiments. The sample configuration in the ESR cell is a bulk plug of millimeter dimensions whereas the light spike samples are only 150-μm films. This suggests that severe radiation trapping and subsequent nonradiative relaxation may be occurring in the ESR cell as opposed to a more optically thin film in this experiment.

Clearly, more experimental work is needed to scan the entire accessible IR region to specifically locate and hopefully identify what frequencies are dominant during a light spike. Future experiments will also attempt to decouple any possible involvement of the sapphire-containment sphere and or impurity-contamination contributions in the sample.

III. CONCLUSION

We have observed light pulses under similar conditions used to generate heat spikes in tritiated solid hydrogen.[4] We believe the decay of the light pulse is independent of the apparatus but possibly a function of the way the sample is distributed on the walls of the sample cell. These optical pulses provide a new method for the study of the dynamics of radiation-induced defects in solid hydrogen. By understanding the recombination process and the steps leading to the recombination spike we may be able to create a bottleneck and increase the atomic hydrogen concentration in the solid or gas phase.

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