

BRIEF REPORTS

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Trapped electrons in solid deuterium

G. W. Collins, P. C. Souers, F. Magnotta, and E. R. Mapoles
Lawrence Livermore National Laboratory, Livermore, California 94551

J. R. Gaines
Department of Physics, University of Hawaii, Honolulu, Hawaii 96822
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We report a narrow electron-spin-resonance (ESR) signal near the free-electron frequency in solid deuterium containing 2% tritium. We speculate that this narrow ESR line is due to localized electrons in bubbles.

We have located a new electron-spin-resonance (ESR) signal from solid deuterium containing 2% of the radioactive isotope tritium. We speculate that this signal is due to electrons trapped in bubbles. Electron bubbles have previously been postulated to explain two broad optical absorptions in irradiated solid hydrogen or deuterium.¹⁻⁴

The experimental details have been described previously.⁵ The spectra were taken under unmodulated adiabatic slow passage conditions at 9.4 GHz. The magnetic field values and absolute electron spin concentrations were referenced to a calibrated ruby standard. The absolute electron concentration is accurate to about 50%, whereas the relative concentration of electrons to atoms is accurate to about 15%. The absolute magnetic field values are accurate to about 8 G while the relative magnetic field values are accurate to within 1 G. The radioactive decay of tritium (T), produces the atoms and electrons described in this paper.⁶ The T_2 used for these experiments was desorbed from a palladium hydride source vessel giving a typical composition of 2% HT, 1% DT, and 97% T_2 . Research grade D_2 was purchased and mixed with T_2 . The sample purity for each experiment was checked up to mass 40 with a Varian MAT Model CH5 magnetic sector mass spectrometer. No evidence of contamination was found in the experiments reported here. After the gas mixture was prepared, the sample was cooled from room temperature, through the triple point (T_{tp}) to 1.4 K. It took our cryostat ~ 20 min to cool from T_{tp} to 1.4 K.

From the top, Fig. 1(a) shows the ESR signal for the middle line of the D atom hyperfine triplet in solid

$D_2 + 2\% T_2$, at 4, 30, and 54 min after cooling to 1.4 K. The zero-field mark is within a few G, of the free-electron value. The doublet structure in the center D resonance is not seen in the other two hyperfine split D lines. By comparison with the other D lines, the left peak in Fig. 1(a) is due to the middle line of the D atom ESR spectrum. The right peak, denoted as E_1 , is a new feature not previously reported. This experiment was repeated twice, once with and once without liquid ^4He placed on the sample for thermal contact.

After the sample sat at the steady temperature of 1.4 K for 23.5 h there was a "heat spike."⁷ The bottom line shape in Fig. 1(a) was collected 26 min after this heat spike.⁸ Figure 1(a) shows the atom and E_1 concentrations increase with time constants that are the same order of magnitude. Also, both the E_1 and atom concentrations decrease simultaneously during a heat spike. After a heat spike, both species grow back as shown in Fig. 1(a). The ESR line shape in Fig. 1(b) shows the same middle D signal after ~ 500 min at 1.4 K when no heat spikes were observed. We see here the narrow E_1 line is lost under the middle D atom line, which at this time in the experiment, resembles the two outside hyperfine split D signals.

From Fig. 1(a), we also find the E_1 linewidth is roughly 25% smaller than the D atom linewidth. After 1 h at 1.4 K, the linewidth (the full width at half maximum peak height) for D atoms⁵ is about 6.2 MHz (2.2 ± 0.2 G) and the E_1 linewidth is about 4.8 MHz (1.7 ± 0.3 G); this sample contains 110 ppm D atoms, 32% $J=1$ concentration, and 25

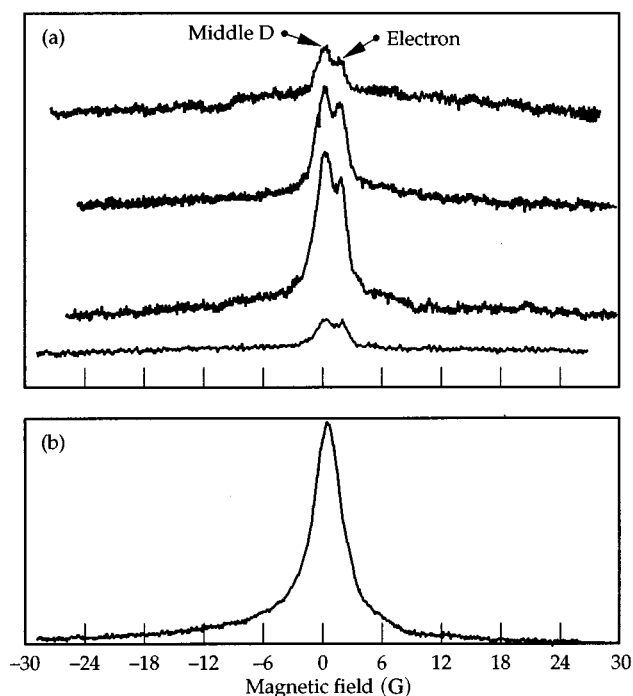


FIG. 1. ESR signal of the middle line of the D atom hyperfine triplet in solid $D_2 + 2\% T_2$ showing the presence of a new narrow line signal just to the high field side of the D line at 1.4 K. (a) From top to bottom, the E_1 and middle D atom line shape at 4, 30, and 54 min after cooling to 1.4 K, and 26 min after a heat spike. (b) The middle D signal at 1.4 K when no heat spikes are observed for ~ 500 min.

ppm E_1 spins. The $J=1$ concentration was calculated from the $J=1$ to 0 conversion time constant of 30 h in $D_2 + 2\% T_2$ at about 1.4 K.⁹

We searched for the E_1 ESR transition in several other hydrogen samples containing tritium. It did not appear in $D_2 + 2\% T_2$ at 4.2 K. We did detect a small narrow line signal near the free-electron frequency in $H_2 + 2\% T_2$ held at 1.7 K, but the signal-to-noise ratio was too small to make quantitative comparisons. There was no doublet feature re-

solved for the middle D atom line shape in $HD + 2\% T_2$ or $D-T$ ($25\% T_2 + 50\% DT + 25\% D_2$) possibly because the linewidths were too broad. We have reported that the middle D atom signal is larger than either of the two outside hyperfine split signals.

The only previous observation of a narrow ESR line near the free-electron frequency in irradiated solid hydrogen comes from Leach's Ph.D. thesis.¹⁰ He studied H atoms in electron-irradiated solid H_2 using rapid passage ESR techniques and found a resonance near the magnetic field value expected for free electrons when he added liquid helium to his samples. He was worried that the signal might be from helium, so he changed from ^4He to ^3He and obtained the same result. The concentration of these spins was much larger below the lambda point of helium than above, suggesting that the thermal conduction of helium was needed to stabilize a large enough concentration for observation. At 1.4 K, Leach measured about 50 times more H atoms than electrons. The linewidth for this signal was ~ 0.5 G which is again $\sim 25\%$ smaller than the 0.6–0.7 G linewidth reported at the same temperature for the H atoms. Thus, the resonant frequency, the temperature dependence, and the relative linewidth for this ESR signal is nearly the same as the E_1 species in D_2 .

To conclude, we have observed a new ESR signal near the free-electron frequency from $D_2 + 2\% T_2$ at 1.4 K. This signal has a narrower linewidth than the linewidth from atoms in the same sample. We speculate that this new signal is caused by an electron bubble, whose radius is larger than the distance between an atom and nearest-neighbor molecule. Both electron bubbles and unpaired atoms disappear during heat spikes.

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⁸The line shape collected after the heat spike is slightly different than the line shape collected at the beginning of the experiment. Due to the limited data we cannot state the significance of this difference.

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