Abstract

In this paper we identify the three THz frequencies that have been found to trigger anomalous exothermy in palladium deuteride with low-lying dipole transition in the deuteron vibrational spectrum. We then speculate that the mesoscopic entanglement that characterizes the ground state of the Tavis-Cummings hamiltonian may give rise to appreciable deuteron-deuteron fusion by virtue of the generic mechanism posited by Takahashi.

On the three exothermic response frequencies when PdD is subjected to coherent THz radiation

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1 Introduction

In a series of really quite remarkable experiments, Letts and Hagelstein [1] succeeded in triggering an anomalous exothermy in Pd saturated with deuterons by means laser irradiation at three specific infra-red frequencies in the range 8 - 25 THz. The frequency selectivity (Q-value) of the anomalies were in the range of 20-40, suggesting a lightly damped and narrow-band resonance response of the target material. Hagelstein has proposed various explanations for the exothermy based on the fact that the lowest of the response frequencies corresponds to the optical phonon band excitation energy, but he has been unable to account for the other (stronger) responses at the higher frequencies.

We are unaware of any other credible attempts to explain this remarkable phenomenon and attribute this to a general scepticism regarding the calorimetric data that we certainly do not share. In this paper we draw attenton to the fact that all of the observed exothermic response frequencies correspond closely to low-lying dipolar transitions in the deuteronic vibration spectrum. We then observe that when the laser radiation is of sufficient intensity all of the deuterons are mutually entangled in a long-lived Dicke ground state. Such a giant-oscillator state constitutes a condensation of the type hypothesized by Takahashi [2]. According to the analysis presented in [2], the rate of nuclear fusion between neighbouring interstitial deuterons is then enhanced by many orders of magnitude over its normal utterly negligible value.

2 Laser-deuteron interaction Hamiltonian

We are going to make several simplifying assumptions. The first of these is that the laser radiation frequency ω excites transitions between a single pair of states in the deuteron vibrational spectrum (with $|E_i - E_j| = \hbar\omega$) and that the plane of polarization is along one of the fcc axes, which we will label x.

According to the Drude model [3], the complex refractive index of Pd at THz frequencies is given approximately by:

$$\epsilon = 1 - \frac{\omega_P^2}{\gamma^2 + \omega^2} + \frac{\omega_P^2 \gamma}{\omega(\gamma^2 + \omega^2)} i$$

implying

$$\sqrt{2} n = \sqrt{|\epsilon| + \mathrm{Re}\{\epsilon\}} + \sqrt{|\epsilon| - \mathrm{Re}\{\epsilon\}} \; i$$

Using $\hbar\omega_P = 2.77eV$ and $\hbar\gamma = 0.068eV$ as published in [4], n at 10 Thz $\approx 24 + 43i$. This means that the laser radiation only penetrates to a depth of around one micron.

Within this surface layer, we envisage N deuterons each occupying one of N interstitial sites of octahedral symmetry. An assembly of two-state quasi atoms driven resonantly by a field of coherent radiation is most simply modelled by the Tavis-Cummings Hamiltonian in which the 2^N state space is transformed into that of a single quasi-spin multiplet. Following [5] and [6] we have

$$H_0 = \hbar\omega a^\dagger a + \hbar\omega \left[\frac{N}{2} + S_3\right] + g(a^\dagger S_+ + a S_-) \tag{1}$$

The sum of the number of photons and excited sites is a conserved quantity (N_{exc}) and the energy levels are given by

$$E_j = N_{exc}\hbar\omega + 2jg\sqrt{N_{exc}}$$

where $-N/2 \le j \le N/2$

The lowest energy is clearly $E_{-N/2} = N_{exc}\hbar\omega - Ng\sqrt{N_{exc}}$ and the gap between levels is $\Delta E = g\sqrt{N_{exc}}$

The field amplitude E_0 of a single photon over N sites each of volume Ω is given by

$$\hbar\omega = 2n^2\epsilon_0 E_0^2 N\Omega$$

where n is the real part of the refractive index at ω.
In a surface layer of depth $\lambda/2$, the coupling to the E-field will vary strongly with position, being greatest for sites at the centre of the layer, but the mean photon-site coupling is approximately described by

$$g_{ij}^2 = e^2 E_0^2 | < i|x|j > |^2 = \frac{e^2 | < i|x|j > |^2 \hbar\omega}{2Nn^2\Omega\epsilon_0}$$

3 Palladium deuteride

Palladium is one of several transition metals that can reversibly absorb hydrogen up to the point of stoichiometry, in which every available interstitial site of octahedral (O) symmetry - is occupied by a hydrogen nucleus. As pointed out in the foregoing introduction, the large mass ratio between the hydrogen nucleus and the host atoms, means that the H quantum oscillation is decoupled from the motion of host atoms and the interstitial hydrogen nucleus experiences an effective static potential, with local mimina at the octahedral and tetragonal symmetry points. Furthermore, each deuteron is screened by the Fermi gas such that intersite deuteron-deuteron interactions are small. The screened deuterons are effectively almost neutral particles, resulting in high mobility within the lattice and a very shallow effective potential. As a consequence, even the ground state has a substantial spatial extent on the order of 0.2Å.

A single deuteron in such an environment exhibits a spectrum of singlet, doublet and triplet state representations of the local point symmetry group. The ground state is a singlet with even parity about the minimum potential along each of the symmetry axes. The next level is a triplet of states with odd parity along one of the symmetry axes and an excitation energy of the order of 0.04 eV ($\approx$ 10 THz). The real and imaginary parts of the refractive index are comparable in this part of the spectrum, so there is effectively only one internally reflected mode along each of the x,y and z axes. and a single photon is effectively normalized to a slab volume $A\lambda/2$. We conclude that microlayers of non-stoichiometric PdD are amenable to the Tavis-Cummings model in which a single photon mode is coupled to N identical two-level quasi-atoms.
In PdD, $\Omega \approx 1.6 \times 10^{-29} m^3$, so

$$\frac{e^2}{\epsilon_0 \Omega} \approx 1.1 \times 10^{21} eVm^{-2}$$

and the dipole transition coupling coefficient is given by:

$$g_{ij}^2 \approx \frac{\hbar\omega_{ij} | < i|x|j > |^2}{Nn^2} \times 5.5 \times 10^{20}\ eVm^{-2}$$

We chose the x axis as our dipole and E-field direction, but exactly the same results would be obtained for y and z axes by virtue of the fcc point symmetry at the octahedral site. μ_x dipoles can only be formed by products of states that have opposite x-parity and equal y and z parities. None of the products involving $[.+-]$,$[.-+]$,$[.--]$ formed x-dipoles any larger than the $[.++]$ parity states and they lie at higher energies, so we do not need to consider them. We solved the $[.++]$ deuteron states on a 3-dimensional grid of pitch 0.25 Å using the semi-empirical effective potential published in [7] and were able to substantially confirm the findings of that paper. The energy levels E_i (relative to the potential minimum at the octahedral symmetry point) of the ten lowest states in each of the [+++] and [-++] subgroups are listed below (in units of meV):

i,j	$E_{i_{[+++]}}$	$E_{j_{[-++]}}$
0	**47**	**88**
1	138	173
2	**190**	190
3	217	**213**
4	**148**	248
5	**148**	182
6	233	283
7	233	283
8	283	**277**
9	284	289

[+++] states {4,5} and {6,7} are degenerate doublets. None of lowest 10 [-++] states are degenerate
We then calculated the μ_x dipole elements. The largest ones are tabulated below, together with the dimensionless photon mode coupling constants $g_{ij}\sqrt{N}/(\hbar\omega)$:

$i_{[+++]}$	$j_{[+--]}$	$\lvert E_i - E_j\rvert \times 10^{-3}$ eV	= THz	E_{lower}	$\langle i\rvert\, x\, \lvert j\rangle$ (Å)	$g_{ij}\sqrt{N}/(\hbar\omega)$
0	**0**	**40**	**9.8**	**47**	**0.16**	**0.08**
1	3	74	18.0	138	0.10	0.07
2	**8**	**87**	**21.1**	**190**	**0.08**	**0.07**
3	1	45	10.8	173	0.17	0.09
3	6	66	15.9	217	0.12	0.08
4+5	**3**	**65**	**15.7**	**148**	**0.19**	**0.11**
6	5	51	12.4	182	0.14	0.08
6	9	56	13.6	233	0.15	0.09
7	5	51	12.4	182	0.14	0.08
7	9	56	13.6	233	0.15	0.09
9	3	72	17.3	213	0.12	0.09

Because the [+++] parity states 4 and 5 are degenerate, we have added their dipole products with [-++] state 3 in quadrature.
The 0-0 transition corresponds to the bottom of the optical phonon band. Comparison with Fig 10b of [1] leads us to make the tentative identifications (shown in **bold**) of the three responses observed at 8.3,15.1 and 20.5 THz with the theoretical dipole transitions at 9.8,15,7 and 21.1 THz. Our second tabulated entry also predicts a strong dipole transition at 18.0 Thz that is not visible in the fitted curve of Fig 10b of [1]. However the raw data in their Table 1 does indeed show an observed exothermic response at 18.0 Thz, which was evidently overlooked in the curve fitting.

4 Enhanced deuteron-deuteron fusion rate

The lowest eigenstate of the Tavis-Cummings hamiltonian is a singlet superposition comprising contributions from every one of the 2^N state of the N quasi-atoms. For example, for $N = 4$ with 4 total excitations we have

$$\psi_{-2} = -0.38\,|0000\rangle + 0.31\,(|0001\rangle + |0010\rangle + |0100\rangle + |1000\rangle) - 0.24\,(|0011\rangle + |0101\rangle + |0110\rangle + |1001\rangle + |1010\rangle + |1100\rangle) + 0.17\,(|0111\rangle + |1011\rangle + |1101\rangle + |1110\rangle) - 0.11\,|1111\rangle \quad (2)$$

The energy gap to the next lowest level (an N-tuplet) is $\sqrt{N_{exc}/2}g\hbar\omega$. We conclude that if the laser radiation is sufficiently strong, the thermodynamically favoured state of the N deuterons is one in which they are completely entangled. Takahashi has investigated [2] the probability that deuterons in such frozen states fuse to form helium-4 and found that the Coulomb barrier is effectively surmounted, leading to appreciable exothermy so long as the entanglement is sustained.

References

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