CHEMISTRY IN LASERS. XIV. UTILIZATION OF NOBLE GAS HYDRIDE LASERS IN THE DISSIPATIVE CONFINEMENT OF TWO-STEP NUCLEAR FUSION OF DOUBLY ISOTOPIC BORON HYDRIDES

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The article is a continuation of the non-classical aspects of fusion (Part XI.). The utilization of doubly isotopic boron hydrides (H—2 and H—3 combined with B—11) in a two-step dissipatively controlled nuclear fusion process is proposed for energy generation, with tandem laser/e-beam pumping. To broaden the optical excitation capability, noble gas hydrides are shown to have principal advantages: lack of a bound ground electronic state (comparable with excimers), far less corrosiveness than noble gas halides (or fluorine containing laser media), wide range of tunability including the soft X-ray/VUV region, and lack of decomposition of the compounds involved upon prolonged usage (superior to dyes).

Two main experimental approaches are available to achieve controlled nuclear fusion, which in turn offers useful energy release: the charged particle accelerator, and thermonuclear fusion. Considerations of a charged particle accelerator are not stressed in this paper, but it is useful to point out the essential contrast between it and thermonuclear fusion: accelerated particles comprise a rather uniform vectorial ensemble with respect to both direction and energy, whereas in the thermonuclear approach the ensemble is normally characterized by a wide distribution of energies as well as random motion. It may also be recalled that it is necessary to distinguish, in the thermonuclear approach, between the kinetic temperature related to Maxwellian distribution, and the equivalent radiation temperature based on black body radiation; the significant differences between the two temperatures can, in the broad sense, provide the perultimate challenge that has attracted many leading scientists to fusion research in recent years.

From a practical standpoint confinement of the fusion reaction center is of paramount importance, and the magnetohydrodynamic approach seems to have taken up a perhaps disproportionate amount of available funds, leaving inertial confinement following laser excitation in the uncertain category. It is important, nevertheless, to discuss the future trend of departing from gas dynamic ablation mo-

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dels in using laser excitation. Up to 50% of the laser energy is transferable into pellett kinetic energy by dynamic absorption and superefficient compression, as was shown by HORE [1] in 1976. The key concept in Hore's approach is to enhance cold implosion. Once cold implosion has passed a critical stage, the system is far enough from equilibrium to be subject to that peculiar branch of non-equilibrium thermodynamics that has gained due recognition recently through the outstanding efforts of Professor Ilva PRIGOGINE and his co-workers, in particular P. GLANSDORFF, R. LEFEVER, and G. NICOLIS. Through autocatalysis or feed-back interaction the non-linear system can form an orderly assembly of new macroscopic dimensions. (The non-symmetrical design of the joint European TORUS may also facilitate such factors in confinement.) The utilization of such "dissipative structures" (popularized by the Zhabotinskieffect) for controlled nuclear fusion has been mentioned previously [2], and now we wish to propose a specific multi-step process that can provide feed-back interaction for confined, oscillatory energy release. Doubly isotopic boron hydrides (H-2 and H-3 combined with B-11), besides offering both hydrogen and boron nuclear fusion in the same molecular dimension, are fascinating chemicals on their own right. Since the first extended report [3] in 1933 on the class of compounds known as boron hydrides, some of the most exciting developments in chemistry have been tied to this area of research. Perhaps the most widely appreciated peculiarity of boron chemistry is the "electron deficiency" of some compounds like B₂H₈, or icosahedral fragments; this has led to the three-center bond concept [4-6] which enjoys general acceptance today. The charge distribution can be approximated in a simple manner from atom-atom polarizability $(\Pi(k, l))$ as:

$$\Pi(k,l) = 2 \sum_{j=1}^{m} \sum_{i=m+1}^{n} \frac{C_{ij}C_{li}}{\varepsilon_i - \varepsilon_i} [C_{kj}(SC)_{ki} + C_{ki}(SC)_{ki}],$$
(1)

where ε_i represent zeroth-order energies, and all levels are empty except those below n, which are doubly occupied. It is of particular interest to us that negative charges tend to be localized in the inner regions of the molecule, and in most cases with disruption of the molecular geometry. This suggests the possibility of highly inelastic interactions with e-beams, due to the availability of very long duration reaction channels (*i.e.*, molecular rearrangement). Some of these rearrangements are also significant in terms of the resurgence of chemical topology in the last few years; the pioneering work of early investigators [7] has never been extended to high energy density regions which are now of fundamental concern, and it is hoped that some effort in basic research will be devoted to this area of boron chemistry in the near future.

Nuclear fusion reactions often occur with greater ease than predicted, *i.e.* there is a discrepancy between the Coulomb barrier calculated according to classical theory, and the experimentally observed barrier penetration (which accounts for the fact that nuclear reactions do occur at detectable rates far below the Coulomb barrier, and provides a powerful argument in favor of quantum mechanics and tunnelling.) Concerning the first of the two fusion steps, it would be counterproductive to attempt to review here the development of hydrogen isotope fusion, an area of chemistry so well established that even elementary texts of recent vintage cover it in considerable detail [8]. Summarizing some of the heavy hydrogen reactions, including the "neutron

branch" (Eq. (2)), the "proton branch" (Eq. (3)), and two reactions (Eqs. (4) and (5)) of high energy yield:

$$D+D \rightarrow He^3 + n$$
 (2)

(0.82 MeV) (2.45 MeV)

 $D+D \rightarrow T +H$ (3) (1.01 MeV) (3.02 MeV)

 $D+T \rightarrow He^4 + n$ (4)

(3.5 MeV) (14.1 MeV) $D + He^3 \rightarrow He^4 + H$ (5) (3.6 MeV) (14.7 MeV)

we find that about 33% of the energy liberated is carried by charged particles to be deposited internally within the reacting system for a pure deuterium target, with the neutrons making additional contributions. This energy would partially aid the next stage: fusion involving boron nuclei.

Compared to the hydrogen isotopes', boron fusion is a new and little explored area. The complete-fusion cross section of boron-11+terbium-159 was reported by KOZUB *et al.* [9] in 1974, while ZEBELMAN and co-workers reported [10] in the same year both fission and completefusion values. Evidence of an intermedate quasi-stationary state of boron-11 fusion was presented [11] by PETKOV *et al.* in 1975, and related [by the same authors to fission calculations in a separate publication [12.] While only a fraction of the boron fusion effort has been fully reported in the open literature, several important advances are documented for the year 1976: (a) DAYRAS, STOCKSTAD, SWITKOWSKI and WIELAND at CIT determined fusion cross sections for $B + {}^{12}C$ over a 5 MeV range extending below the Coulomb barrier, and compared the results with optical model calculations [13]; (b) NAMBOODIRI and cp-workers examined fusion products of ${}^{12}C + {}^{12}C$, and considered the boron cross section too [14]; (c) DATLA *et al.* used a small θ -pinch plasma to deduce ionization rate coefficients for B(IV) as well as C(V) [15].

Among the large number of possible compounds, B_4H_{10} and B_5H_{11} are typical suitable analogs, with varying atomic ratios of the desired isotopes. In the tandem excitation process laser driven heavy hydrogen fusion is the initiator for the boron-11 fusion, which is e-beam pumped. The energetic feed-back leading to oscillatory confinement of the reaction center need not necessarily be tied to a classical reaction of nuclear chemistry, but could be due, for example, to self-focussing control of the energy release process as a function of dissipative structures. To aid the oscillatory mechanism, the energy input is of crucial importance, and several aspects can only be decibed by experiment. In the following we address ourselves to the question of upgrading laser performance.

In the quest for suitable materials to provide high output power, tunability, short wavelength, and if possible exceptional stability, various shortcomings of available laser systems become readily apparent. By resorting to the concept of strongly coupling the radiation field in an optical resonant cavity with a suitable

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molecular ensemble [16], the feasibility of preparing some optimal compounds in situ became an evident route of novel significance. Hydrogen, the simplest chemical element, is an ideal counterpart to noble gases, due to the lack of a stable hydride with any of them. It is instructive to pursue the details of thermodynamic stability vs. instability based on known bond energetics, and the excellent and provoking text by DASENT [17] should be consulted by those unfamiliar with the field. The necessary orbital overlap for hydride formation can be produced by hyperpolarization-with varying facility as a function of the number of electrons involved, the extent of the electron cloud and its polarizability, hence as a function of atomic number or isotope in case of hydrogen. To select energy levels one needs to refer, perhaps paradoxically, not to general texts on quantum chemistry, but to Kittel's "Thermal Physics" [18]. The crucial trade-off of ordinary molecular orbitals for the alternate set available in the OR cavity is readily acceptable once an essentially relativistic treatment, based on photon-cavity interactions rather than the atomic and molecular electronic transition lines, is adopted. The soundness of the approach can be gauged from theoretical [19] as well as experimental grounds; the latter stems from Stanford University's spectacular development of a free-electron laser (yet to be published), in which a relativistic electron beam creates the energy levels for population inversion through interaction with a pulsed magnetic field.

Additional factors influencing tuneability are the overall pressure, temperature, and above all: composition. With several noble gases in the cavity, staircase tuning to short wavelengths is the obvious approach, hydride formation giving great flexibility in comparison to noble gas excimers/exciplexes only. Unlike CO_2 (which has been successfully tuned already by controlling added inert gas components and overall pressure), the decomposition products in the present system are innocuous starting materials. A TEA arrangement is promising in view of VUV laser action having been reported in pure hydrogen upon electric discharge. A similar arrangement reported in 1973 [20], based on electric discharge pumping of noble gas-scintillator dye vapor mixtures in an optical cavity, was subject to dye decomposition and slow coating of the mirrors from the cavity side—complications which are, of course, totally eliminated when rare gases and hydrogen comprise the optically active medium.

References

- [1] Hore, H.: Proc. Eur Electro-Opt. Mark. Technol. Conf. (2nd). (1976).
- [2] Keszthelyi, C. P.: Chemistry and Lasers 3, 641 (1976).
- [3] Stock, A.: "Hydrides of Boron and Silicon", Cornell University Press, Ithaca, New York, 1933.
- [4] Stitt, F.: J. Chem. Phys. 8, 891 (1940).
- [5] Pitzer, K. S.; J. Amer. Chem. Soc. 67, 1126 (1946).
- [6] Mulliken, R. S.: Chem. Rev. 41, 207 (1947).
- [7] Dickerson, R. E., W. N. Lipscomb: J. Chem. Phys. 27, 212 (1957).
- [8] Gordon, G., W. Zoller: "Chemistry in Modern Perspective", Addison Wesley, 1973.
- [9] Kozub, R. L., D. Logan, J. M. Miller, and M. Zebelman: Phys. Rev. C 10, 214 (1974).
- [10] Zebelman, A. M., L. Kowalski, J. Miller, K. Beg, Y. Eyal, G. Jaffe, A. Kandil, and D. Logan: Phys. Rev. C 10, 200 (1974).
- [11] Petkov, I., Ya. Delchev: Bulg. J. Phys. 2, 293 (1975).
- [12] Delchev, Ya., I. Petkov, and Kh. Khristov: Yad. Energ. 2, 13 (1975).
- [13] Dayras, R. A., R. G. Stokstad, Z. E. Switkowski, and R. M. Wieland: Nucl. Phys. A A261 478 (1976).
- [14] Namboodiri, M. N., E. T. Chulick, and J. B. Natowitz: Nucl. Phys. A A263, 491 (1976).

[15] Datla, R. U., L. J. Nugent, and Hans R. Griem: Phys. Rev. A 14, 979 (1976).

[16] Keszthelyi, C. P.: Spec. Ltrs. 3, 85 (1974).

[17] Dasent, W. E.: "Nonexistent Compounds", Marcel-Dekker, Inc., New York, 1965.

[18] Kittel, C.: "Thermal Physics", John Wiley and Sons, Inc., New York, 1969.
[19] Wigner, E. P.: Rev. Mod. Phys. 29, 255 (1975).

[20] Keszthelvi, C. P.: Ph. D. Thesis, University of Texas at Austin, 1973.

ЛАЗЕРНАЯ ХИМИЯ, XIV. ПРИМЕНЕНИЕ ЛАЗЕРОВ НА ИНЕРТНЫХ ГАЗОВЫХ ГИДРИДАХ В ДИССИПАТИВНЫХ СТАБИЛИЗАЦИЯХ ДВУХСТУПЕНЧАТОЙ ЯДЕРНОЙ ФУЗИИ ДВОЙНЫХ ИЗОТОПОВ БОРОГИДРИДОВ

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Настоящая статья является продолжением серии статей по неклассическим аспектам ядерной фузии (часть XI.). Предлагается применение двойных изотопов борогидридов (H-2 и H-3 в сочетании с B-II) в двухступенчатых контролируемых ядерных фузионных процессах для генерации энергии с помощью накачки электронного пучка тандемного лазера. Для расширения области оптической возбуждаемости, гидриды инертных газов обладают следующими принципиальными преимуществами: 1. отсуствие связного основного электронного состояния (подобно эксимерам); 2. гораздо менее коррозионные чем галиды инертных газов (или, чем лазерные материалы содержащие фтор); 3. широкая область подстройки, включая мягкие рентгеновские лучи и вакуумного ультрафиолета; 4. отсуствие разложения вещесть участвующих в продолжительной эксплоатации лазера (превосходя красители).