Atomization of correlated molecular-hydrogen chain: A fully microscopic Variational Monte-Carlo solution

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Ustroń, September 11, 2018

Outline

R. P. Dias, I. F. Silvera, Science 10.1126/science.aal1579 (2017)

The New York Times


Mount Etna, Europe's Most Active Volcano, Puts On a Show


In California, a Move to Ease the Pressures on Aging Dams


Edward E. David Jr., Who Elevated Science Under Nixon, Dies at 92


Before Vaquitas Vanish, a Desperate Bid to Save Them

SCIENCE

Hydrogen Squeezed Into a Metal, Possibly Solid, Harvard Physicists Say

By KENNETH CHANG JAN. 26, 2017

Frankfurter Allgemeine

Wissen

RUBRIKEN MEDIZIN GENE KLIMA WELTRAUM GARTEN NETZRÄTSEL

MATERIALFORSCHUNG EXTREM

Wasserstoff zu Metall gequetscht?

VON MANFRED LINDINGER

Ingeniøren

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Metallisk hydrogen sætter forskerverdenen i kog

Påstand om fremstilling af metallisk hydrogen mødes med meget hård kritik fra forskere. Lige til skraldepanden, lyder det. Andre bakker dog de kritiserede forskere op.

Af Jens Ramskov 2. feb 2017 kl. 12:03

ВЕСТИ.RU Новости Видео Фотолента Трансляции Сервисы

Тема: Сверхпроводники и изучение сверхпроводимости

Новости | Наука

30 января 2017 15:00 | Дарья Загорская

Прорыв в физике? Твёрдый металлический водород, возможно, стал реальностью

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INNOVATION

Scientific breakthrough lost? Unique metallic hydrogen sample disappears

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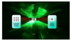
31 gennaio 2017

Idrogeno solido metallico, un annuncio e molti dubbi


Credit: Ranga P. Dias/Isaac F. Silvera

Due ricercatori hanno annunciato di aver prodotto per la prima volta idrogeno solido metallico, previsto per via teorica circa ottant'anni fa, un traguardo che aprirebbe la strada a nuove applicazioni, dai superconduttori ai propellenti per razzi. Ma non pochi scienziati nutrono dubbi riguardo alle modalità con cui è stato svolto l'esperimento e dunque al suo risultato (red)

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World's first metallic hydrogen sample disappears

Last month physicists from Harvard University in the US had claimed to have successfully turned hydrogen into a metal - something researchers had been struggling to achieve for more than 80 years.

PTI | Posted by Bijin Jose
Boston, February 27, 2017 | UPDATED 19:20 IST

INDEPENDENT

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News › Science

World's only piece of a metal that could revolutionise technology has disappeared, scientists reveal

Exclusive: Harvard University physicists say first-ever piece of metallic hydrogen on Earth has been lost after catastrophic failure of diamond holding it under enormous pressure

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REUTERS

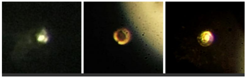
EDITION: INDIA

Business Markets India World Tech Commentary Breakingviews Money Sport & Life

TECHNOLOGY NEWS | Fri Jan 27, 2017 | 10:02am IST

U.S. scientists create metallic hydrogen, a possible superconductor, ending quest

FULL COVERAGE: INDIA ELECTIONS 2017



RMF 24 NAJBLIŻEJ FAKTÓW

FAKTY OPINIE AKCJE RMF FM ROZRYWKA ZDJĘCIA FILMY

RMF 24 • Fakty • Nauka • Metaliczny wodór, materiał marzeń, stał się rzeczywistością

Metaliczny wodór, materiał marzeń, stał się rzeczywistością

Czwartek, 26 stycznia (23:40)

Jego istnienie fizycy przewidywali od 80 lat. Teraz wreszcie stał się faktem. Naukowcy z Uniwersytetu Harvarda ogłosili właśnie, że udało im się stworzyć metaliczny wodór, materiał o potencjalnie rewolucyjnych właściwościach. Na razie jego wytworzenie wymaga skrajnie niskiej temperatury i olbrzymiego ciśnienia, większego, niż w samym środku Ziemi, jeśli okazałby się stabilny w normalnych warunkach, mógłby być w temperaturze pokojowej nadprzewodnikiem. To oznaczałoby rewolucję w wielu dziedzinach, choćby związanych z przesyłem energii. Pisze o tym w najnowszym numerze czasopismo "Science".

Metalization of Hydrogen

Metalic state

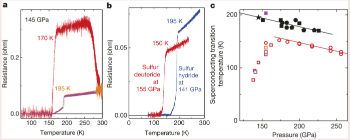
E. Wigner i H. B. Huntington, J. Chem. Phys. **3**, 764 (1935):

- $H-H$ distance (d_{HH}),
- Wigner-Seitz radius ($r_s \equiv (\frac{3}{4\pi n})^{1/3}$).

Metalization at $p \approx 25 GPa$:

$$2r_s > d_{HH}.$$

Hydrogen in 2D - superconductivity?



A. P. Drozdov et al., Nature **525**, 73 (2015)

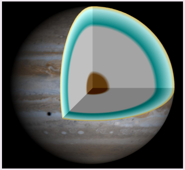
Superconductor

N. Ashcroft, PRL **21**, 1748 (1968)

$$T_C = \Theta_D \mathcal{F}(\lambda(r_s))$$

- Θ_D - Debye Temperature,
- λ - electron-phonon coupling.

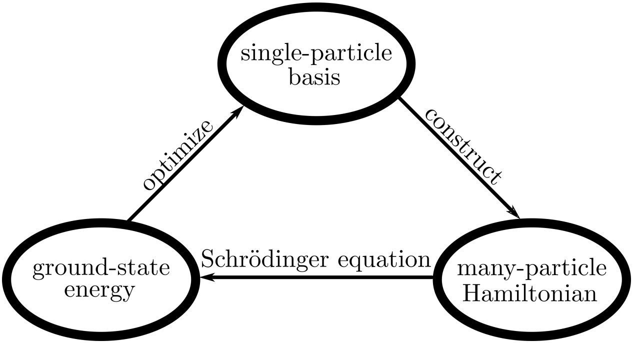
	r_s (a_0)	T_C (K)
Jupiter surface	0.1	$2e-27$
Jupiter core	0.8	283.4



en.wikipedia.org/wiki/Metallic_hydrogen

Jupiter core: superconductor with $T_C \sim 300\ K$?

Exact Diagonalization Ab Initio (EDABI) + VMC



N-paticle correlated state: $\left|\Psi^{N}_{trial}\right\rangle \equiv \hat{\mathcal{P}}\left|\Phi_{free}\right\rangle$;

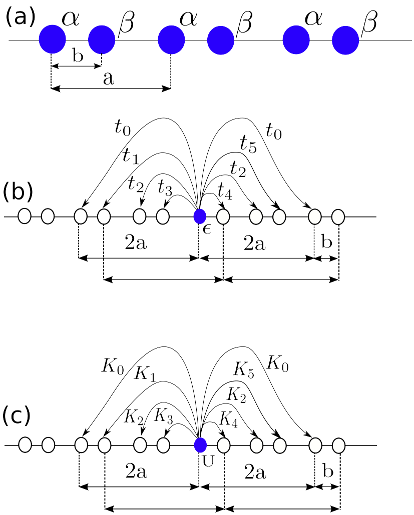
Jastrow correlator: $\hat{\mathcal{P}} = e^{-\sum_{i\neq j}\lambda_{i,j}\hat{n}_i\hat{n}_j - \sum_i \lambda^U_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}}$;

Observables: $\langle\hat{\mathcal{O}}\rangle \equiv \frac{\left\langle\Psi^N_T\middle|\hat{\mathcal{O}}\middle|\Psi^N_T\right\rangle}{\left\langle\Psi^N_T\middle|\Psi^N_T\right\rangle}$.

Sources

♠ J. Spałek et al., Phys. Rev. B 61, 15676 (2000); ♣ APK et al., Eur. Phys. J. B 86, 252 (2013);
♦ A. Biborski, APK, J. Spałek, Comput. Phys. Commun. 197, 7 (2015);
♡ A. Biborski, APK, J. Spałek, Phys. Rev. B 98, 085112 (2018).

For details see: Phys. Rev. B **98**, 085112 (2018)



Assumptions

(a) two hydorgen atoms in the unit cell (α, β), with the lattice parameter *a* and bond length *b*;
(b) range of the hoppings terms extends up to 2*a*;
(c) interactions counted up to the range of 2*a*.

"Infinite" crystal

- Periodic Boundary Conditions;
- supercell of 17, 21, 25, 33 and 37 unit cells;

Hamiltonian

Second quantization

$$\mathcal{H} = \sum_i \epsilon_i (\hat{n}_{i\uparrow} + \hat{n}_{i\downarrow}) + \sum_{i \neq j} t_{ij} (\hat{c}^\dagger_{i\uparrow} \hat{c}_{j\uparrow} + \hat{c}^\dagger_{i\downarrow} \hat{c}_{j\downarrow}) \qquad \text{// free electrons}$$

$$+ \sum_i U_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + \sum_{i \neq j} K_{ij} \hat{n}_i \hat{n}_j \qquad \text{// interactions}$$

First-to-second-quantization calculation step

$$t_{ij} \equiv \left\langle w_i(\mathbf{r}) \middle| -\nabla^2 - \textstyle\sum_{l \in \text{ions}} \frac{2Z}{|\mathbf{R}_l - \mathbf{r}|} \middle| w_j(\mathbf{r}) \right\rangle \qquad \epsilon_i \equiv t_{ii}$$

$$V_{ijkl} \equiv \left\langle w_i(\mathbf{r}) w_j(\mathbf{r}') \middle| \frac{2}{|\mathbf{r} - \mathbf{r}'|} \middle| w_k(\mathbf{r}) w_l(\mathbf{r}') \right\rangle \qquad U_i \equiv V_{iiii}, \; K_{ij} \equiv V_{ijij}$$

Dimensionality - 1D chain in 3D space

- $w_i(\mathbf{r})$ build from 1s Slater orbitals;
- Coulomb potential $V_C(\mathbf{R}) \propto |\mathbf{R}|^{-1}$;

Proper state function

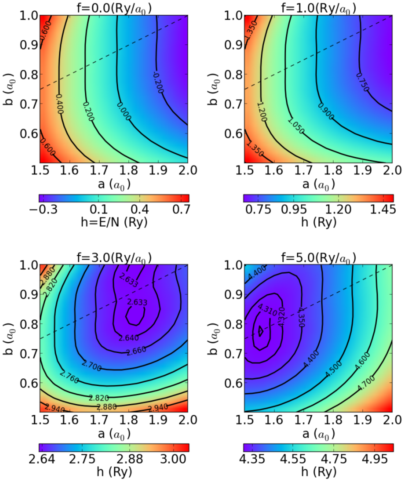
One-dimensional enthalpy

$$h \equiv f\frac{a}{2} + \frac{E}{N},$$

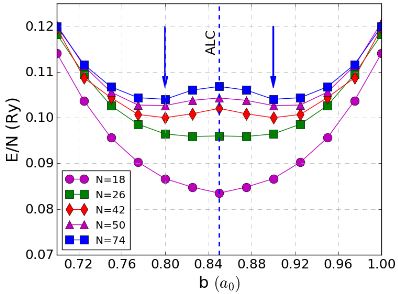
with f as an external force (analogue of the pressure), the lattice parameter a, and ground-state energy E for the N-particle supercell.

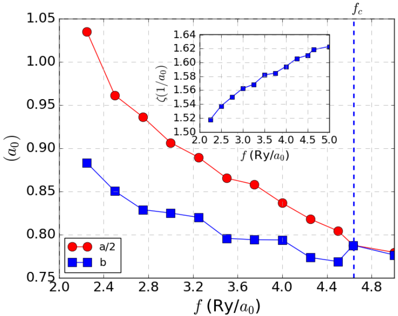
Run for given f

- opt. structure
  - opt. wavefunction
    - opt. Jastrow



Results for finite systems





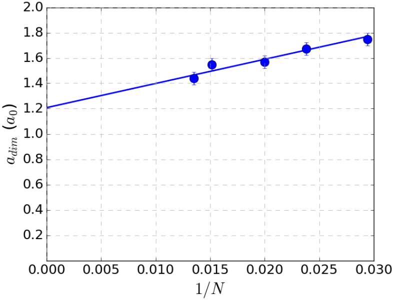
Peierls-like distortion from First Principles for a correlated system

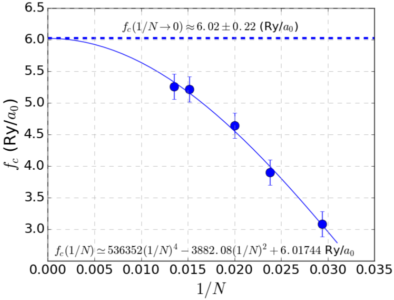
- **No distortion for small systems.**
- Molecular $\rightarrow$ atomic transition at high "pressure"

$\hookrightarrow$ see my poster after lunch.

for finite systems cf. also E. Giner *et al.*, J. Chem. Phys. **138**, 074315 (2013).

Thermodynamic limit





Conditions of molecular-to-atomic transition for $N \to \infty$

- finite-size scaling of atomization lattice parameter $a_{dim} \approx 1.17 a_0 \; > \; 0$;
- finite-size scaling of atomization force $f_c \approx 6.02 \frac{Ry}{a_0} \; < \infty$.

Metallicity of hydrogen chain

Point of reference

We use the parameters of the Hamiltonian for $N = 50$ for the reference.

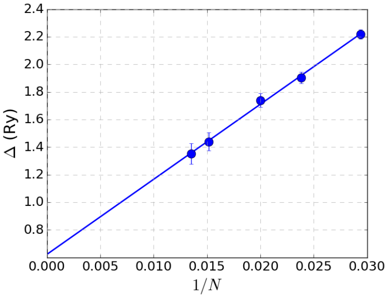
Charge gap

$$\Delta_N \equiv \left.\frac{E_{N+4} - 2E_N + E_{N-4}}{4}\right|_{@h(f)}$$

E_N - the ground state of the N-particle system described by the reference Hamiltonian with the structure minimizing effective enthalpy.

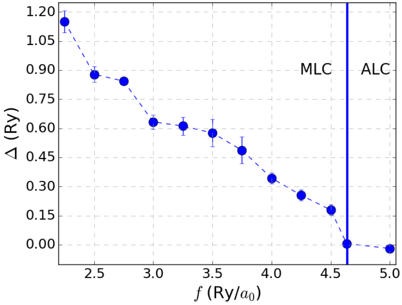
Thermodynamic limit

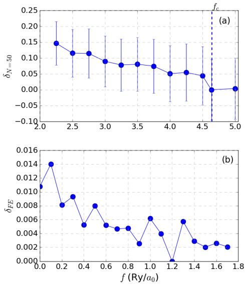
$$\Delta \equiv \Delta_\infty = \lim_{N\to\infty} \Delta_N$$



Example of finite-size scaling for charge gap Δ.

Closing of the charge gap





Apparent metallicity of the hydrogen chain in the atomic phase

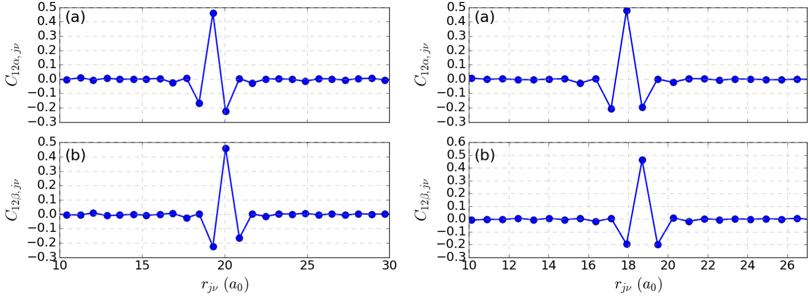
- charge gap closed at the MLC $\rightarrow$ ALC transition;
- further-than-nearest neighbor hoppings;
- chain exist in 3D (both single-particle wavefunctions and Coulomb potential are taken for $D = 3$);

in agreement with L. Stella *et al.*, Phys. Rev. B **84**, 245117 (2011)

Density-density correlation

Density-density correlation

$$C_{i,j} \equiv \langle \hat{n}_i \hat{n}_j \rangle - \langle \hat{n}_i \rangle \langle \hat{n}_j \rangle$$

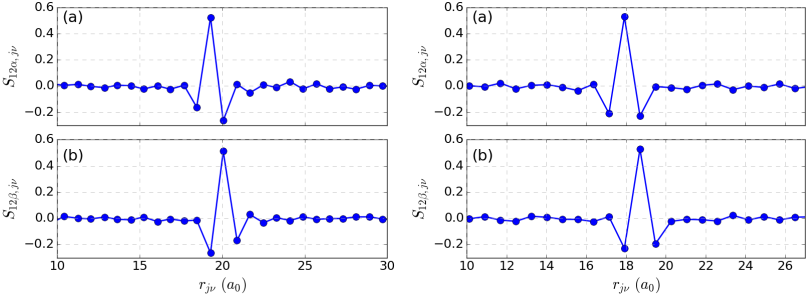


(LEFT) $f = 4.5$ Ry$/a_0$; (RIGHT) $f = 5.0$ Ry$/a_0$

Spin-spin correlation

Spin-spin correlation

$$S_{i,j} \equiv \left\langle (\hat{n}_{i\uparrow} - \hat{n}_{i\sigma})(\hat{n}_{j\uparrow} - \hat{n}_{j\sigma}) \right\rangle = \langle \hat{S}_i^z \hat{S}_j^z \rangle$$

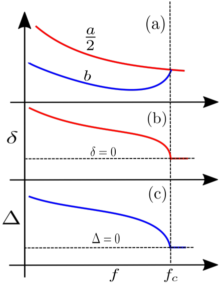


(LEFT) $f = 4.5$ Ry$/a_0$; (RIGHT) $f = 5.0$ Ry$/a_0$

Conclusions

Hydrogen chain

- Peierls-like distortion at ambient "pressure";
- correlations do not weaken distortion;
- external force induces molecular $\rightarrow$ atomic transition;
- concomitant atomization and metallization ;
- no long-range order;



Dziękuję za uwagę   Thank you for your attention

See me at the poster session to hear about Hydrogen in 2D