First-principle approach to correlated realistic molecular hydrogen planes: Role of the Heisenberg-type interaction and the superconductivity

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NARODOWE CENTRUM NAUKI

Zakopane, September 21, 2018

Outline

1. Motivation
   - Media frenzy
   - Hydrogen under pressure
2. Methods
   - EDABI + VMC
   - Model
3. Results
   - Transition sequence
   - Metallicity
   - Superconductivity
4. Conclusions

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

Wasserstoff zu Metall gequetscht?

Ingeniøren

Nyheder Blogs Debat Jobfinder Avisen Mere

VORES FOKUS | KUNSTIG INTELLIGENS | 3D-PRINT | DIESELSKANDALEN | KAMPFLY FOR MILLI

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

Тема: Сверхпроводники и изучение сверхпроводимости

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

FOX NEWS Tech

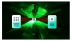
Scientific breakthrough lost? Unique metallic hydrogen sample disappears

Le Scienze

EDIZIONE ITALIANA DI SCIENTIFIC AMERICAN

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

indiatoday NEWS TV

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

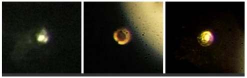
INDEPENDENT

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

REUTERS

TECHNOLOGY NEWS | Fri Jan 27, 2017

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



RMF 24 NAJBLIŻEJ FAKTÓW

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 czasopisma "Science".

Hydrogen under pressure

TH: 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 $p \approx 25 GPa$:

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

TH: Superconductivity in $300K$ (?)

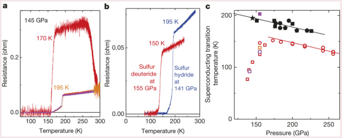
N. Ashcroft, PRL **21**, 1748 (1968)

$$T_C = \Theta_D \mathcal{F}(\text{el.-ph.})$$

	T_C (K)
Jupiter surface	$\sim 10^{-27}$
Jupiter core	~ 290

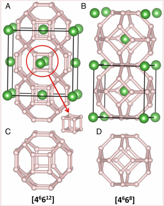
Hydrogen in 2D - superconductivity?

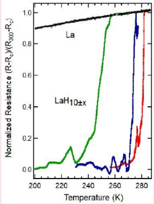
$H_{2|3}S$



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

$LaH_{10} \pm x$

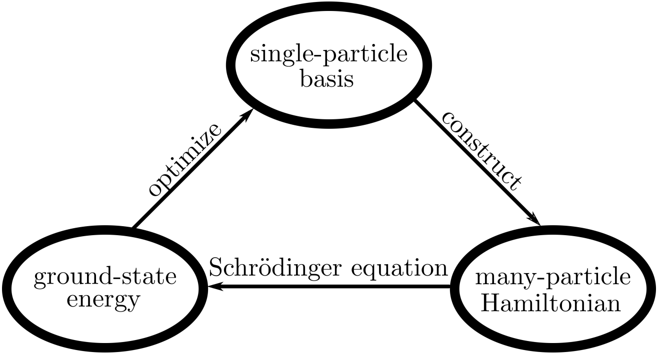




L$_{\text{theory}}$: Hanyu Liu et al., PNAS **114**, 27 (2017)

R$_{\text{experiment}}$: M. Somayazulu et al., arXiv:1808.07695 (2018)

Exact Diagonalization **Ab** Initio (EDABI)++

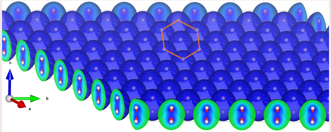


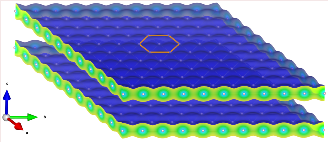
Sources

♠ 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).
Coming soon: EDABI for *f* electrons..

Triangular lattice

Two-dimensional crystal





- periodic boundary conditions in xy plane;
- Lanczos algorithm for the diagonalization core of 6 and 8 atoms (to comply with proper Néel 120° and 90° phases);
- wavefunction constructed from 10 classes of nodes

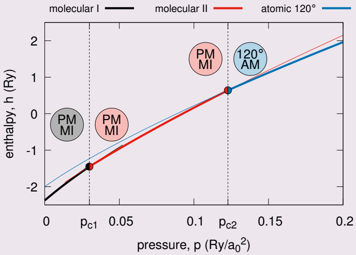
$$\mathcal{H} = \sum_{i\sigma} \epsilon_i \hat{n}_{i\sigma} + \sum_{i\neq j\sigma} t_{ij} \hat{c}^{\dagger}_{i\sigma} \hat{c}_{i\sigma}$$ ↪ hoppings t_{ij} up to 10^{th} neighbor;

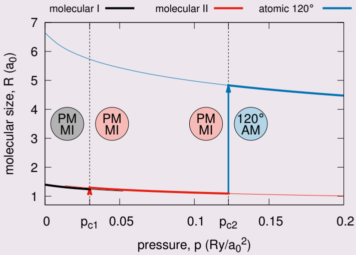
$$+ \sum_i U_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow} + \sum_{i\neq j} K_{ij} \hat{n}_i \hat{n}_j$$ ↪ Coulomb repulsion K_{ij} up to 10^{th} neighbor;

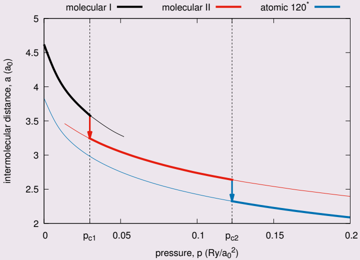
$$- \sum_{i\neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} \sum_{i\neq j} J_{ij} \hat{n}_i \hat{n}_j$$ ↪ ferromagnetic exchange J_{ij} up to 3^{rd} neighbor;

$$+ \sum_{i\neq j} J_{ij} \hat{c}^{\dagger}_{i\uparrow} \hat{c}^{\dagger}_{i\downarrow} \hat{c}_{j\downarrow} \hat{c}_{j\uparrow}$$

2D enthalpy and lattice parameters







Question:

What is the quantum equivalent of $R_{eff} \longrightarrow \infty$?

Atomicity

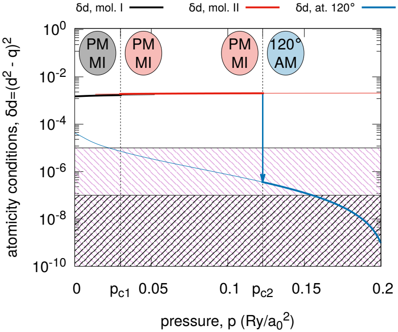
Classically

Interplanar distance $R_{eff} \longrightarrow \infty$ $\Leftarrow$ **Not necessarily in the quantum realm!**
(van-der-Waals-like behavior)

Independence of classical probability

$$\delta d \equiv \left(P\begin{pmatrix} * \\ \uparrow\downarrow \end{pmatrix} P\begin{pmatrix} \uparrow\downarrow \\ * \end{pmatrix} - P\begin{pmatrix} \uparrow\downarrow \\ \uparrow\downarrow \end{pmatrix} \right)^2$$

$$\equiv \left(\langle \Phi_0 | \hat{n}_{1\uparrow}\hat{n}_{1\downarrow} | \Phi_0 \rangle \langle \Phi_0 | \hat{n}_{2\uparrow}\hat{n}_{2\downarrow} | \Phi_0 \rangle - \langle \Phi_0 | \hat{n}_{1\uparrow}\hat{n}_{1\downarrow}\hat{n}_{2\uparrow}\hat{n}_{2\downarrow} | \Phi_0 \rangle \right)^2$$



Magnetic order

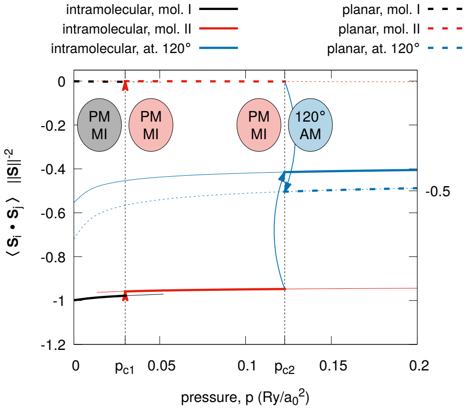
FM vs. AFM exchange

$J_{\text{FM, Hund-like}} \ll J_{\text{AFM, kinetic}}$

Required for the ambient pressure stability of the atomic phase!

Spin correlation

1. Molecular phases: molecular near spin-singlet H_2
2. Atomic phase: 120° Néel order



Total spin

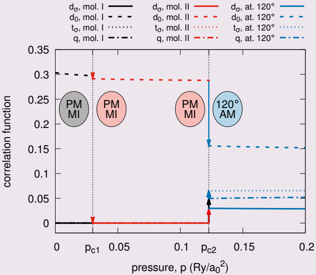
	mol. I → II		mol. II → atomic	
$\|\|S\|\|_{\text{molecule}}$	0.10	0.14	0.16	0.54
$\|\|S\|\|_{\text{triangle}}$	0.86	0.87	0.86	0.077

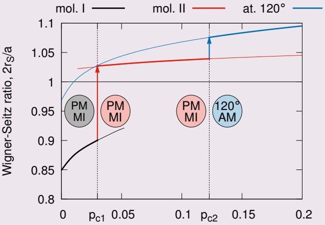
$$||S||_{\textbf{molecule}} \equiv ||S(x_{2D}, -\tfrac{R}{2}) + S_2(x_{2D}, \tfrac{R}{2})||$$

$$||S||_{\textbf{triangle}} \equiv ||S(x_{2D}, \tfrac{R}{2}) + S(x_{2D} + e_1, \tfrac{R}{2}) + S(x_{2D} + e_2, \tfrac{R}{2})||$$

Two-step metallization

Metallicity of atomic phase





$$q \equiv P\begin{pmatrix} \uparrow\downarrow \\ \uparrow\downarrow \end{pmatrix} \qquad d_0 \equiv P\begin{pmatrix} \uparrow & \\ & \downarrow \end{pmatrix}$$

$$t_\uparrow \equiv P\begin{pmatrix} \uparrow & \\ & \uparrow\downarrow \end{pmatrix} \qquad d_\uparrow \equiv P\begin{pmatrix} \uparrow \\ \uparrow \end{pmatrix}$$

$$t_\downarrow \equiv P\begin{pmatrix} & \downarrow \\ \uparrow\downarrow & \end{pmatrix} \qquad d_\downarrow \equiv P\begin{pmatrix} \downarrow \\ \downarrow \end{pmatrix}$$

(top): occupancy correlation functions

(bottom): Wigner-Seitz metallicity condition

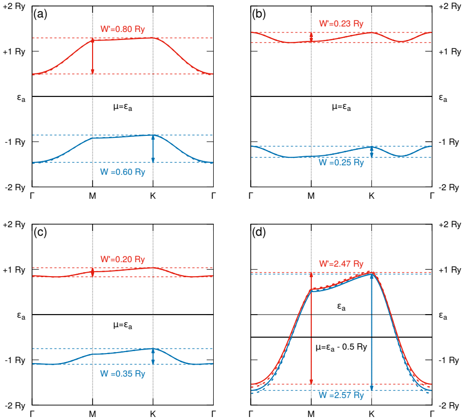
$$r_S \equiv \left(\frac{3}{4\pi n}\right)^{1/3}$$

Band structure

Bare bands
- easily calculable
- depend only on $\mathcal{H}_{\text{free}}$

Correlated bands
- full $\mathcal{H}$ dependence
- no generic method

Bare bands with a correlator
- calculable
- local interaction
- ∅ correlator physics



Possibility of superconducting state

Wigner-Seitz radia

$r_S = r_S(V)$

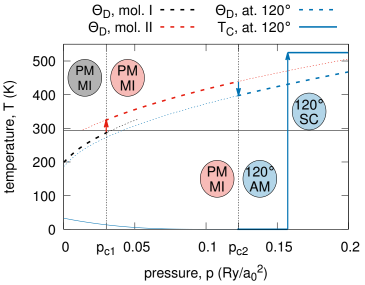
- vol. of an electron in ph. I & II:
  $$V_e = \frac{V_{\mathbf{mol}}}{2} \equiv \frac{1}{2} a^2 (R + \frac{2}{\zeta}),$$
- vol. of an **atom** in atomic phase:
  $$V_e = a^2 \frac{2}{\zeta},$$

source	method	r_s (a_0)
J. McMinis et al. (arXiv:1309.7051)	DMC	2.27
G. Mazzola et al. (Nat. Commun. **5**, 3487 (2014))	DMC	**1.28**
J.-L. Li et al. (Phys. Rev. B **66**, 035102 (2002))	LSDA	2.78
J.-L. Li et al. (Phys. Rev. B **66**, 035102 (2002))	GGA	2.50
B. I. Min et al. (Phys. Rev. B **33**, 324 (1986))	LMTO-LSDA	2.85
A. Svane et al. (Solid State Commun. **76**, 851 (1990))	SIC-LSDA	2.45
B. G. Pfrommer et al. (Phys. Rev. B **58**, 12680 (1998))	GGA-PW91	2.5
APK, AB, JS (2018)	EDABI	**1.265**
R. P. Dias et al. (Science: 10.1126/science.aal1579 (2017))	eksperyment	**1.255 – 1.34**

McMillana formula

T_C depends on

- Θ_D (from phonon spectra)
  - $\varnothing$ always a soft mode $\perp$ to the plane
- $\alpha \approx 1.0$
- $\lambda^2 \approx 0.166 r_S$
  - $\varnothing$ tolerable (?) approx.



Conclusions

Physics of hydrogen planes

- concomitant atomization & metallization;
- long-range interactions ($\sim ||\mathrm{R}||^{-p}$)
- London-like interactions in insulating molecular phases;
- benchmark for infinite-system quantum chemistry (EDABI + QMT);

Hydrogen-induced superconductivity

- medianly correlated system (playground for a physicist)
- (most probably)[citation needed] anharmonic phonons;
- (but maybe)[citation needed] correlation driven;
- extreme pressure (chemical?);
- record high T_C;

Thank you for your attention