Electron-lattice coupling and superconductivity in hydrogen-rich systems

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Kraków, Dec 4, 2019

Outline

1. Motivation
   - Media frenzy
   - Hydrogen under pressure
2. Methods
   - EDABI++
   - Model
3. Results
   - Transition sequence
   - Metallicity
4. Superconductivity
   - Eliashberg Theory
   - Phonons
5. 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 Wissen

MATERIALFORSCHUNG EXTREM

Wasserstoff zu Metall gequetscht?

VON MANFRED LINDINGER

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 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

FOX NEWS Tech

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INNOVATION

Scientific breakthrough lost? Unique

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 (red)

indiatoday.in NEWS TV

INDIA TODAY CONCLAVE 2017 ASSEMBLY ELECTIONS 2017 MAIL TODAY INDIA TODAY

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

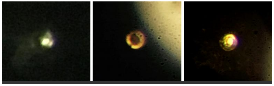
Exclusive: Harvard University physicists say first-ever piece of metallic hydrogen on Earth has been lost after catastrophic failure of diamond holding it under enormous pressure

REUTERS

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

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 być w normalnych warunkach, mógłby być w temperaturze pokojowej nadprzewodnikiem. To oznaczałoby rewolucję w wielu

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 $300K$ (?)

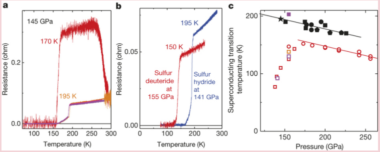
N. Ashcroft, PRL **21**, 1748 (1968)

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

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

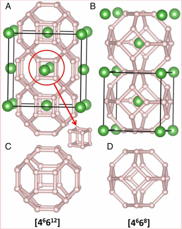
Hydrogen in 2D - superconductivity?

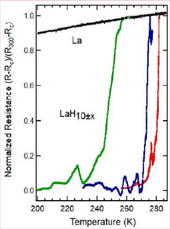
$H_{2|3}S$



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

$LaH_{10} \pm x$

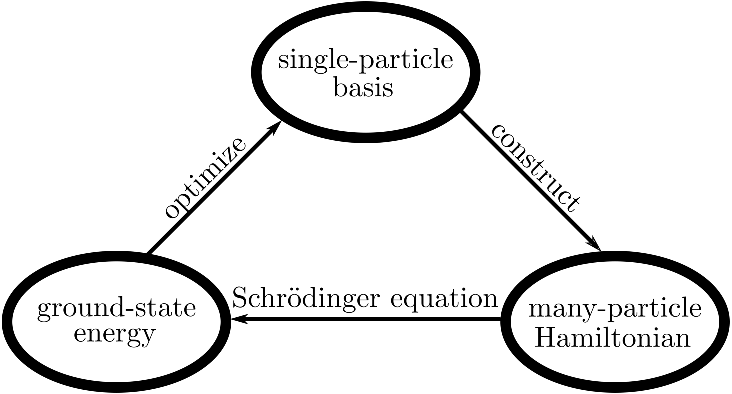




L$_{\text{theory}}$: Hanyu Liu et al., PNAS **114**, 27 (2017)

R$_{\text{experiment}}$: M. Somayazulu et al., PRL **122**, 027001 (2019)

Exact Diagonalization Ab Initio (EDABI)++

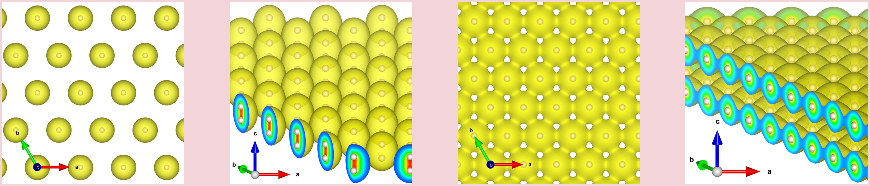


Sources

♠ 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

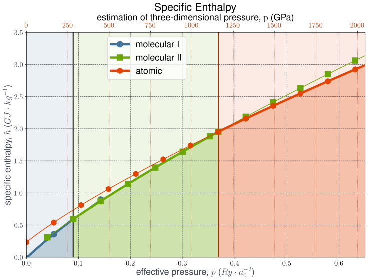
$$\mathcal{H} = \sum_{i\sigma} \epsilon_i \hat{n}_{i\sigma} + \sum_{i\neq j\sigma} t_{ij} \hat{c}^\dagger_{i\sigma} \hat{c}_{i\sigma}$$ $\hookrightarrow$ hoppings t_{ij} up to 10^{th} neighbor;

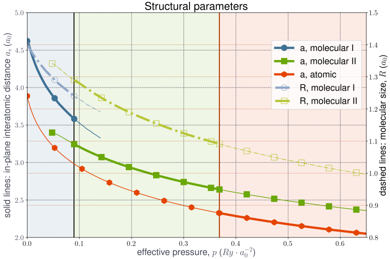
$$+ \sum_i U_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + \sum_{i\neq j} K_{ij} \hat{n}_i \hat{n}_j$$ $\hookrightarrow$ Coulomb repulsion K_{ij} up to 10^{th} neighbor;

$$- \sum_{i\neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} \sum_{i\neq j} J_{ij} \hat{n}_i \hat{n}_j$$ $\hookrightarrow$ ferromagnetic exchange J_{ij}

$$+ \sum_{i\neq j} J_{ij} \hat{c}^\dagger_{i\uparrow} \hat{c}^\dagger_{i\downarrow} \hat{c}_{j\downarrow} \hat{c}_{j\uparrow}$$ up to 3^{rd} neighbor;

2D enthalpy and lattice parameters



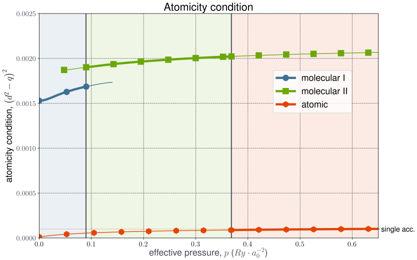


Question:

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

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

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



Magnetic order

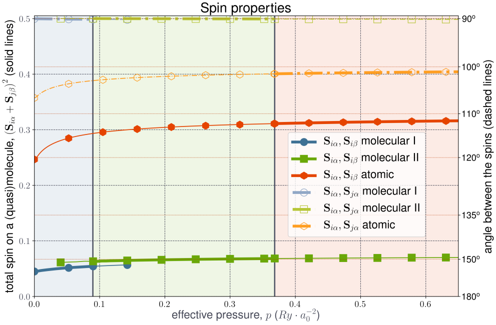
FM vs. AFM exchange

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

Required for the ambient pressure stability of the atomic phase!

Spin correlation

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



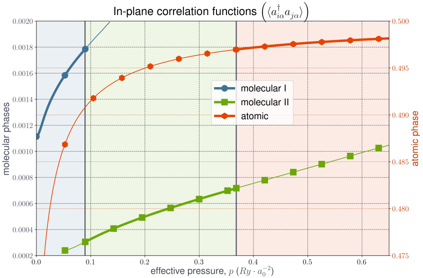
Total spin

	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

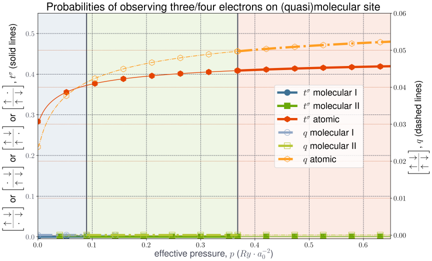
$$||\mathbf{S}||_{\text{molecule}} \equiv ||\mathbf{S}(\mathbf{x}_{2D}, -\tfrac{R}{2}) + \mathbf{S}_2(\mathbf{x}_{2D}, \tfrac{R}{2})||$$

$$||\mathbf{S}||_{\text{triangle}} \equiv ||\mathbf{S}(\mathbf{x}_{2D}, \tfrac{R}{2}) + \mathbf{S}(\mathbf{x}_{2D} + \mathbf{e}_1, \tfrac{R}{2}) + \mathbf{S}(\mathbf{x}_{2D} + \mathbf{e}_2, \tfrac{R}{2})||$$

Metallization I: Correlation Functions



$$\mathcal{C}_{ij} \equiv \left\langle \hat{c}^\dagger_{i\sigma} \hat{c}_{i\sigma} \right\rangle = \left\langle \Phi_0 \middle| \hat{c}^\dagger_{i\sigma} \hat{c}_{i\sigma} \middle| \Phi_0 \right\rangle_G$$

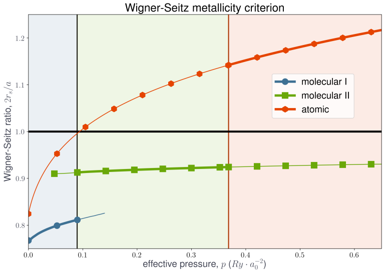


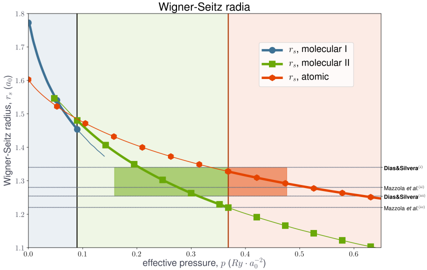
$$q \equiv P\begin{pmatrix} \uparrow\downarrow & \\ \uparrow\downarrow & \end{pmatrix} \qquad d_0 \equiv P\begin{pmatrix} \uparrow & \\ & \downarrow \end{pmatrix}$$

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

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

Metallization II: Wigner-Seitz Criterion





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

$$\text{metal} \Leftrightarrow 2r_s > d_{HH}$$

Can be found experimentally!

source	method	$r_s(a_0)$
Min et al., PRB **33**, 324 (1986)	LMTO	2.85
Pfrommer et al., PRB **58**, 12680 (1998)	GGA-PW91	2.50
Svane et al., SSC **76**, 851 (1990)	LSDA	2.45
Li et al. PRB **66**, 035102 (2002)	LSDA	2.78
Li et al. PRB **66**, 035102 (2002)	PBE	2.50
Mazzola et al., Nat.C. **5**, 3487 (2014)$^{(i)}$	DMC + MD	**1.28**$^{(ii)}$
McMinis et al., arXiv:1309.7051 (2013)	DMC	2.27
AB,APK,JS, PRB **96**, 085101 (2017)$^{(iii)}$	EDABI	**1.27**
molecular II	EDABI	$\mathbf{1.22}^{+0.17}_{-0.06}$
atomic	EDABI	$\mathbf{1.33}^{+0.10}_{-0.04}$
Dias & Silvera	experiment	**1.297(43)**

Metallization III: Band structure

Bare bands

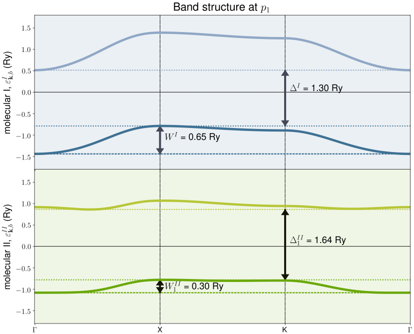
- easily calculable
- depend only on $\mathcal{H}_{\text{free}}$

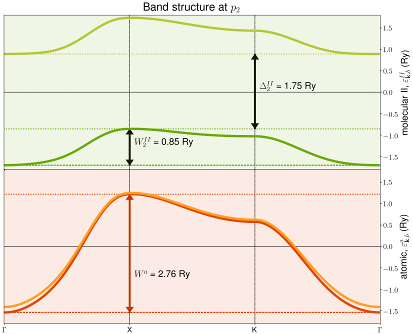
Correlated bands

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

Bands + Correlator

- calculable
- ∅ correlator physics





Possibility of superconducting state

Conventional Superconductivity

Atomic hydrogen is **metallic** ⇔ **McMillan formula** for critical temperature

McMillan formula

$$T_C = \frac{\Theta_D}{1.45} \exp\left[-\frac{1.04(1+\lambda)}{\lambda + \mu^*(1+0.62\lambda)}\right]$$

- Θ_D - Debye temperature (from phonon DOS)
- λ - electron phonon coupling (from phononic and electronic dispersions)
- μ^* - Morel-Anderson pseudopotential - typically fitted to experimental data

We attempt to derive the ab-initio value of pseudopotential μ^*.

Morel-Anderson pseudopotential

$$\mu^* = \frac{\mu}{1 + \mu \log(\frac{\mathrm{T}_{\textbf{phonons}}}{\mathrm{T}_{\textbf{electrons}}})}$$

$$\mu^* = \frac{n(E_F)(U - K_1)}{1 + n(E_F)(U - K_1)\log(\frac{E_f}{k_B\Theta_D})}$$

Electron - phonon coupling

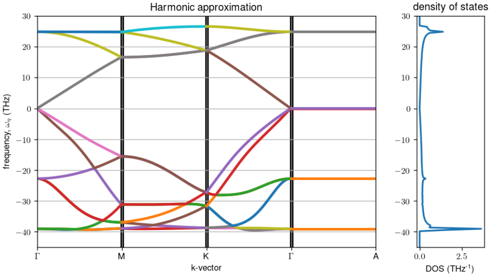
Eliashberg spectral function

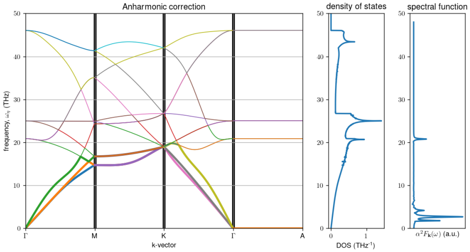
$$\alpha^2 F_{\mathbf{k}}(\omega) \sim \sum_\eta \int d\mathbf{q} M_\eta^2 \delta(\omega - \omega_\eta)\delta(\varepsilon(\mathbf{k}) - \varepsilon(\mathbf{k}+\mathbf{q}))$$

allows us to obtain electron-phonon coupling constant

$$\lambda = 2\int_0^\infty \frac{d\omega}{\omega}\alpha^2 F_{\mathbf{k_f}}(\omega)$$

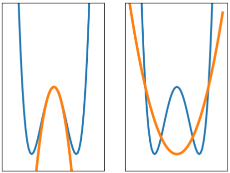
Electrons and Phonons: DFT calculations with EDABI constrains





We take the Mexican-hat potential:

$$U(\{u^i\}) = U_0 + \frac{1}{2}\Phi_{ij}u^{ij} + \frac{1}{4!}\Phi_{ijkl}u^{ijkl}$$



$$\mathbf{F}_i \to \mathbf{F}_i + \frac{1}{4!}\Phi_{i;j\langle kl\rangle}u^{j\langle kl\rangle}.$$

At $p_{\text{eff}} = 0.7\,Rya_0^{-2}$ ($\sim 1\,TPa$)

U_{eff} (Ry)	μ^*	λ
1.194	0.192	1.05
Θ_D (K)	T_C (K)	T_{AD} (K)
1300	164	**176**

SCAN meta-GGA + vdW corrections in DFT calculations

Conclusions

Physics of hydrogen planes

- concomitant atomization & metallization;
- long-range interactions ($\sim ||\mathbf{R}||^{-p}$);
- London-like interactions in insulating molecular phases (true molecular crystal);
- weak London-like attraction of atomic planes;
- benchmark for infinite-system quantum chemistry (EDABI + QMT);

Hydrogen-induced superconductivity

- **medianly** correlated system;
- anharmonic correction to force constants nesesery;
- superconductivity induced by electron-phonon coupling;
- Morel-Anderson pseudopotential **from First Principles**;
- high critical temperature $T_C = 176K$;
- extreme pressure (chemical?);

Thank you for your attention