Foundation for Polish Science

INNOVATIVE ECONOMY
NATIONAL COHESION STRATEGY

EUROPEAN UNION
EUROPEAN REGIONAL
DEVELOPMENT FUND

Autumn School on Correlated Electrons: Emergent Phenomena in Correlated Matter, Jülich 2013

Metallization of Atomic Solid Hydrogen within the extended Hubbard model

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MOTIVATION

One of the important problems in Condensed Matter Physics are the metal-insulator transition of the Mott-Hubbard type [1]. Our aims are to investigate:

1. quantum critical behavior of the wave function near Mott transition;
2. evaluation of the electron wave function in the strongly correlated system;
3. effect of the external magnetic field;
4. combination of first and second quantizations;
5. stabilizing pressure of the crystal near MIT.

As an extra, 5 can be interpreted as a metallization pressure of atomic hydrogen.

We start with the extended Hubbard model [3] at half filling ($\langle n \rangle = 1$):

$$\begin{aligned}\mathcal{H} =& \epsilon_a^{eff} \sum_i n_i + \sum_{i \neq j, \sigma} t_{ij} a_{i\sigma}^{\dagger} a_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \\ &+ \frac{1}{2} \sum_{i \neq j} K_{ij} \delta n_i \delta n_j - \sum_{i,\sigma} \sigma h n_{i\sigma},\end{aligned} \tag{1}$$

where $\delta n = 1 - n$, $h = \frac{1}{2} g \mu_B H_a$ is a reduced magnetic field.

HAMILTONIAN PARAMETERS

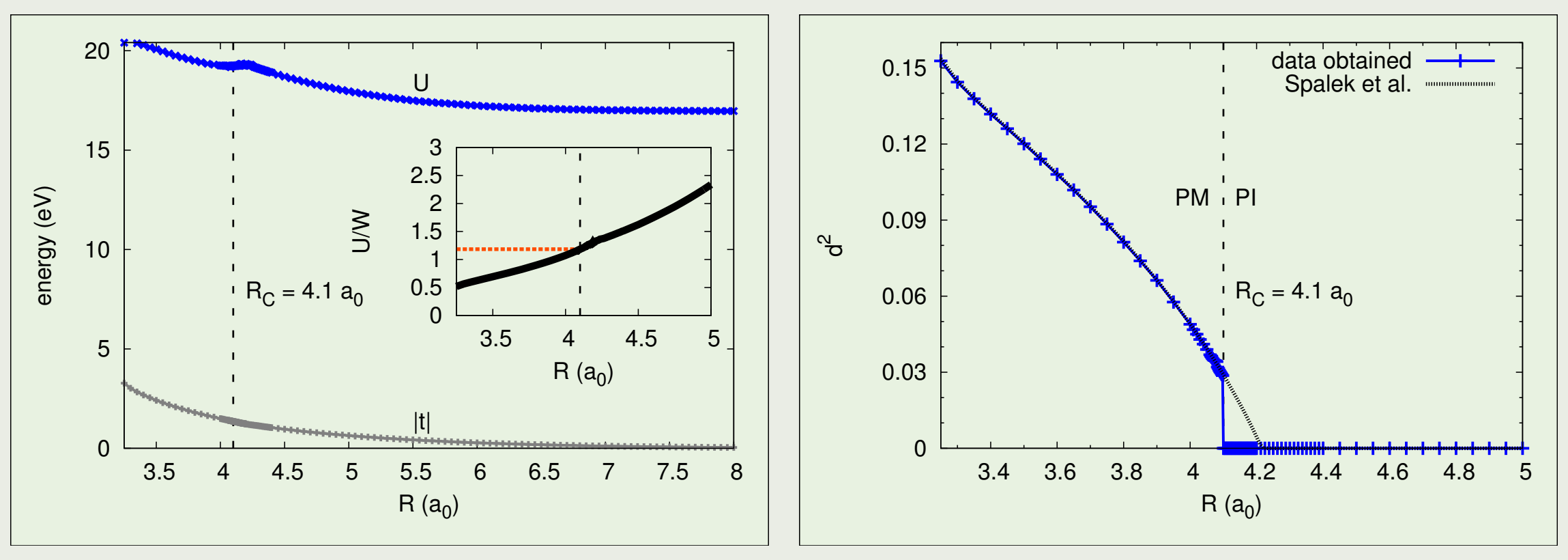

Figure 1 L: Microscopic parameters: hopping integral $|t|$ and the Hubbard interaction parameter U, both as a function of R. Inset: U/W ratio vs R. The U/W ratio for $R = R_c$ is $(U/W)_C \simeq 1.18$. The vertical dashed line marks the $PM \to PI$ transition point. In the large-R limit U reaches the atomic value $U_{at} = (5/4)\,Ry$.
Figure 1 R: Double occupancy probability $d^2 = \langle n_{i\uparrow} n_{i\downarrow} \rangle$ versus R. Note a weak discontinuous jump to zero at $R = R_c \approx 4.1 a_0$, as compared to the continuous evolution for $PM \to PI$ of the Gutzwiller approximation obtained previously [2].

METHOD APPLIED

EDABI

To obtain Hamiltonian parameters we calculate integrals:

$$t_{ij} = \langle w_i | H_1 | w_j \rangle, \tag{2}$$

$$U = \langle w_i w_i | V_{12} | w_i w_i \rangle, \ etc., \tag{3}$$

where H_1 is the Hamiltonian for a single particle, and V_{12} represents interparticle interaction, by approximating Wannier orbital by the series of Gaussian functions:

$$w_i(\underline{r}) = \beta \Psi_i(\underline{r}) - \gamma \sum_{j=1}^{z} \Psi_j(\underline{r}), \tag{4}$$

$$\Psi_i(\underline{r}) = \sqrt{\frac{\alpha^3}{\pi}} e^{-\alpha |\underline{r} - \underline{R_i}|} \approx \alpha^{\frac{3}{2}} \sum_{a=1}^{n} B_a \left(\frac{2\Gamma_a^2}{\pi} \right)^{\frac{3}{4}} e^{-\Gamma_a^2 |\underline{r} - \underline{R_i}|^2}. \tag{5}$$

β and γ parameters depend explicitly on the integrals of Ψ_i functions and z is the number of nearest neighbors. Parameters B_a and Γ_a are derived by minimizing energy of single atom (Hamiltonian $\mathcal{H} \overset{a.u.}{=} -\nabla^2 - \frac{2}{|\underline{r} - \underline{R_i}|}$). n is a number of Gaussian functions used for the approximation. Parameter α is found as a value minimizing the ground energy.

Statistically-Consistent Gutzwiller Approximation (SGA)

To minimize α for each considered system we have to obtain its ground energy. It was proven [4] that Gutzwiller Approximation not always results in finding the lowest energy. Thus we introduce two additional molecular fields λ_m and λ_n, coupled with m and n respectively, and obtain energy of an electron as given

$$E_{\mathbf{k}\sigma} = q_\sigma \epsilon_{\mathbf{k}} - \sigma (h + \lambda_m) - (\mu - \lambda_n), \tag{6}$$

with the bare dispersion relation $\epsilon_{\mathbf{k}}$ and Gutzwiller coefficient q_σ. For a purpose of minimizing energy we minimize functional $\mathcal{F}$:

$$\begin{aligned}\mathcal{F}^{(SGA)} = &-\frac{1}{\beta} \sum_{\underline{k}\sigma} \log \left(1 + e^{-\beta E_{\underline{k}\sigma}^{(SGA)}} \right) \\ &+ \Lambda \left(\lambda_n n + \lambda_m m + U d^2 \right).\end{aligned} \tag{7}$$

GENERAL PROPERTIES

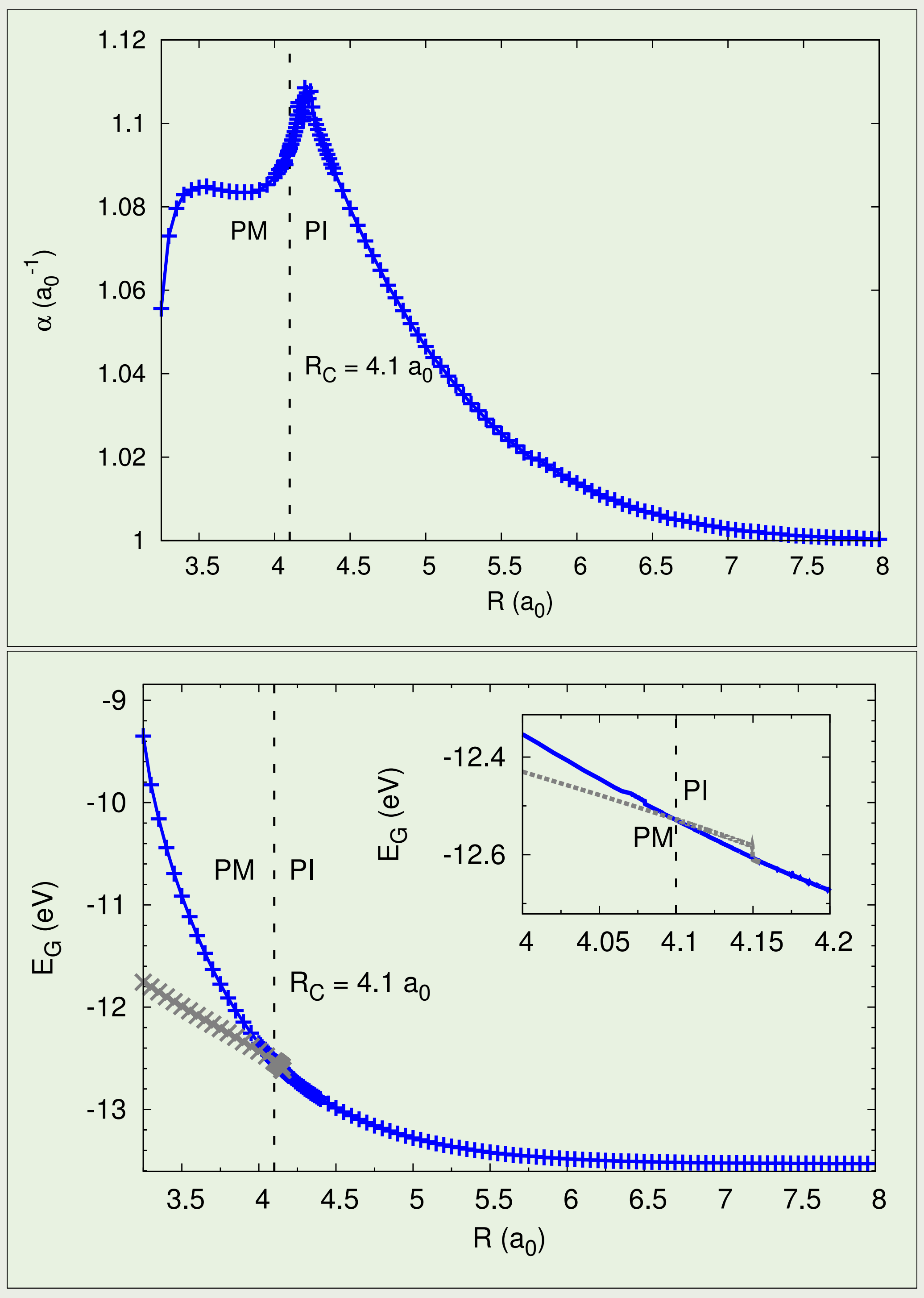

Figure 2 T: Inverse atomic-orbital size vs. R. The critical behavior near $R = R_c = 4.1 a_0$. The dependence is practically independent of the applied field. Note that the cusp has its maximum slightly above R_c.
Figure 2 B: Ground state energy (per atom) of the metallic state $(PM, \times)$ for $R < R_c$ and the insulating $(PI, +)$ for $R \geqslant R_c$, as a function of interatomic distance R. Inset: detailed representation of the first-order $PM \to PI$ transition near $R = R_c \approx 4.1 a_0$. The upper curve for $R < R_c$ represents the energy of the unstable PI state. Note that as $E_G > -1 Ry$, the lattice can only be stabilized by the external pressure.

DETAILED CHARACTERISTICS AT MIT

Calculations were performed on 96-thread node at *ATOMIN Cluster* at Marian Smoluchowski Institute of Physics, for SC 3D crystal (hence $z = 6$) for band filling factor $n = 1$. For optimizing complexity $STO - 3G$ basis (3 Gaussian per Ψ_i) was chosen. Alternative $STO - 7G$ would slightly improve accuracy but the average execution time would be increased by the factor of 30.

GROUND-STATE PROPERTIES

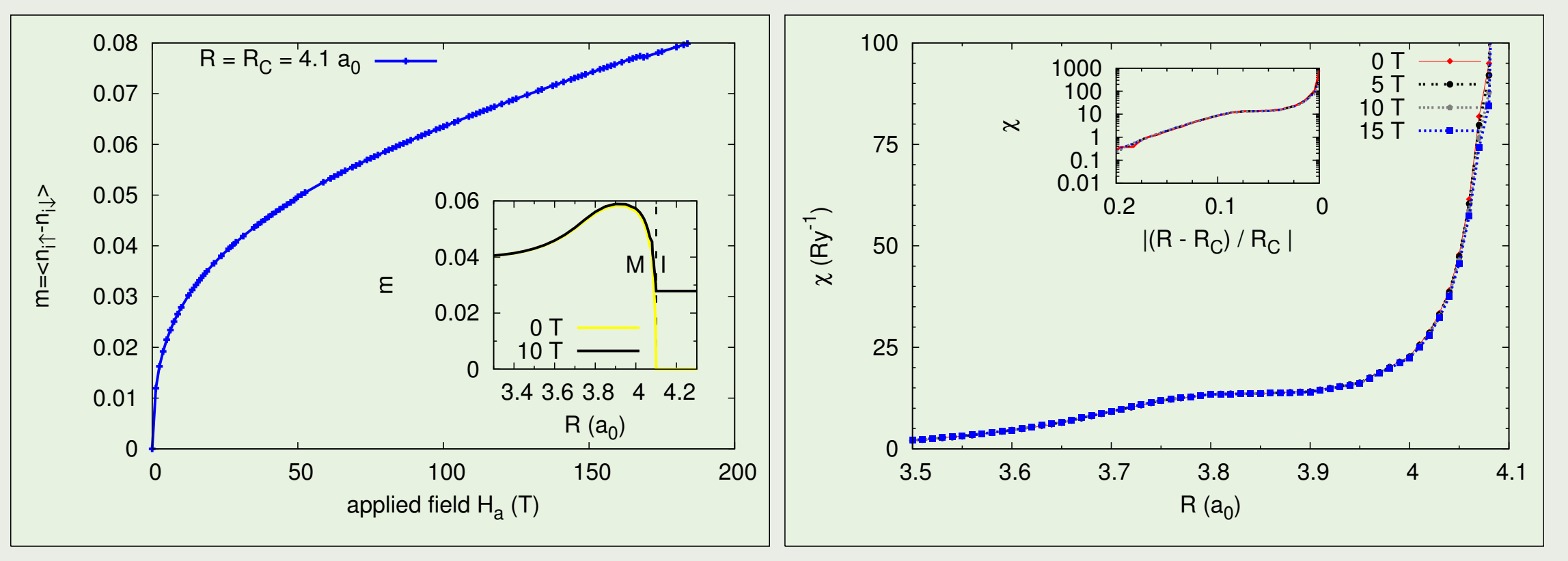

Figure 3 L: Magnetic moment $m = \langle n_{i\uparrow} - n_{i\downarrow} \rangle$ as a function of the applied magnetic field at the critical interatomic distance $R_c = 4.1 a_0$. Inset: a residual uniform moment in the metallic state vs. R near R_c, induced by the correlation field when it is assumed as spatially uniform.
Figure 3 R: Differential static magnetic susceptibility vs. R for selected values of the applied magnetic field. The χ divergence at $R_c = 4.1 a_0$ accompanies the $PM - PI$ transition and is associated with localization of the itinerant electrons when $R \to R_c$. Overall χ behavior in the metallic state does not depend much on the value of H_a. Inset: double logarithmic plot $\chi(R)$ showing absence of any simple exponential type of scaling.

SOLID ATOMIC HYDROGEN: THE CRITICAL PRESSURE FOR METALLIZATION

Our formulation differs from the standard treatment [5], where the phase diagram is a function of microscopic parameter U/W, since we include a procedure of evaluating the renormalized–by–correlations wave functions. Thus we can calculate *explicitly* the critical pressure for metallization.

For such a purpose we calculate pressure p as the force $F = |-\nabla_R E_G|$ applied to stabilize crystal over the area $A = R^2$ of a single cell.

One can observe the obvious discontinuity in pressure at critical lattice parameter $R_C = 4.1 a_0$ caused by the weak (first order) metal–insulator transition (see Figs. 1R and 2B). Hence we can obtain the metallizing critical pressure $p_{C,I} = 97.7 GPa$.

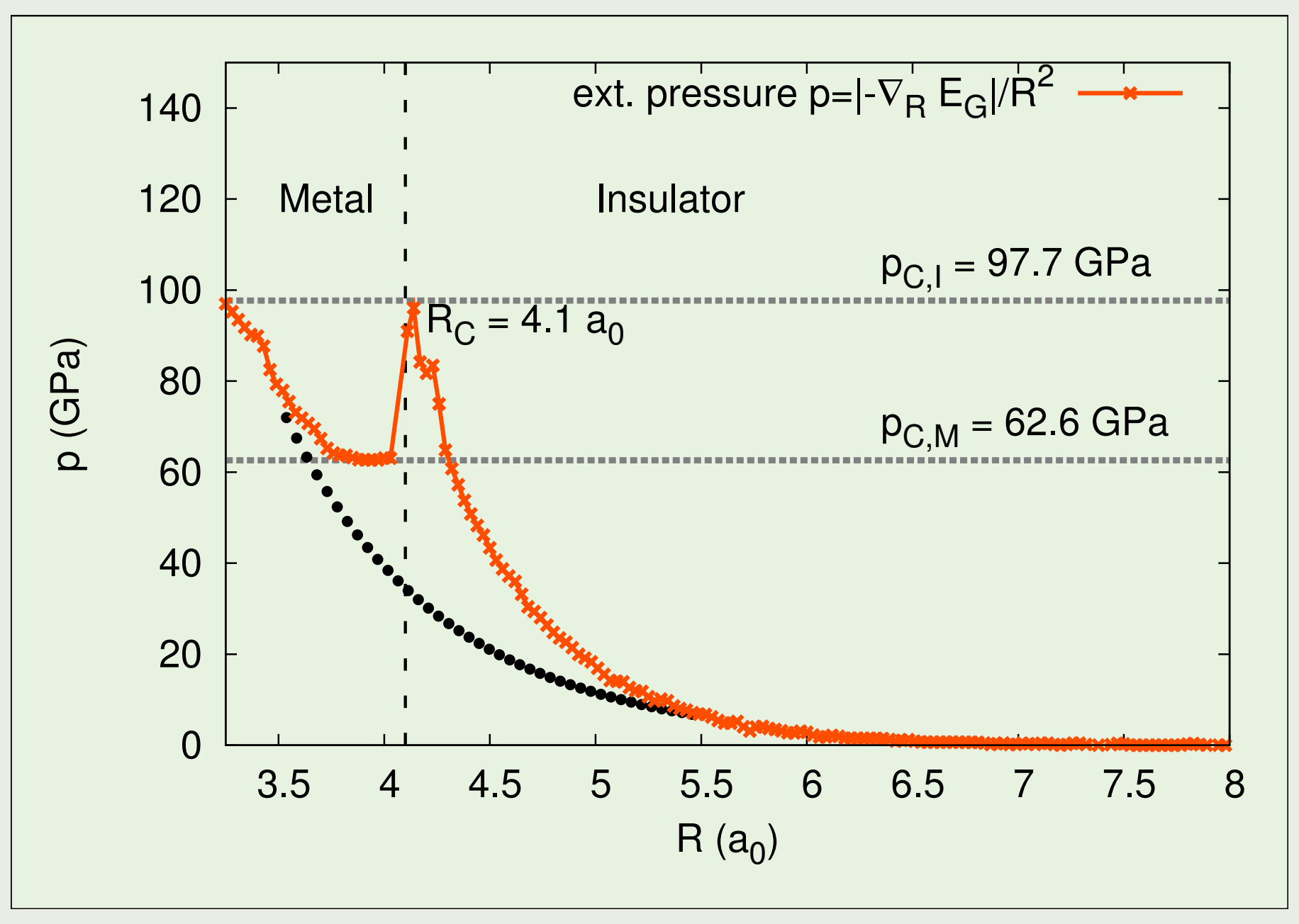

Figure 4: External pressure one has to exert in order to stabilize the crystal vs. interatomic distance (with cell area $A/N = R^2$). Note two critical values of pressure: $p_{C,I} = 97.7 GPa$ required in the insulating state and $p_{C,M} = 62.6 GPa$ in the metallic state. The lattice becomes very rigid as $R \to R_c$. The dotted line marks naive extrapolation $M \to I$.

ACKNOWLEDGMENTS

Discussions with Dr. Jan Kaczmarczyk and Marcin Abram are greatly appreciated. The work was realised in the Project TEAM awarded to our group by the Foundation for Polish Science (FNP) for the years 2011-2014. The partial support by the special Grant MAESTRO from the National Science Center (NCN) for the years 2012-2017 should also be acknowledged.

PUBLICATION

This results has been published in June 2013 at *The European Physical Journal B* issue 86.

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