Dynamical Magnetic Excitations in Nanostructures Deposited on Surfaces

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Abstract

State of the art experimental methods are being developed to investigate magnetic excitations of surfaces[1] or of nanoobjects deposited on surfaces[2]. A key quantity describing these excitations is the transverse dynamical susceptibility, that is a computational burden, accessible from tight-binding methods[3] but very rarely calculated from density functional theory and limited to bulk systems[4].

Using the Korringa-Kohn-Rostoker Green function method within the framework of time dependent density functional theory, we developed a new, efficient and computationally attractive real space method that allows to tackle magnetic excitations in nanostructures. The behavior of adatoms and dimer is investigated..

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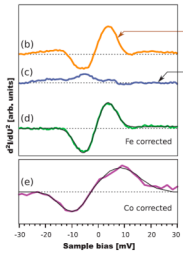
3 R. B. Muniz and D. L. Mills, Phys. Rev. B **68**, 224414 (2003); *ibid.* **66**, 174417 (2002).
4 S. Y. Savrasov, Phys. Rev. Lett. **81**, 2570 (1998); J. B. Staunton *et al.*, Phys. Rev. Lett. **82**, 3340 (1999); P. Buczek et al., Phys. Rev. Lett. **102**, 247206 (2009); E. Sasioglu *et al.*, Phys. Rev. B **81**, 054434 (2010).

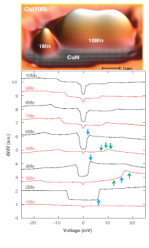
Introduction: Experimental evidence

Fe and Co adatoms on Pt(111)



Balashov et al., PRL 102, 257203 (2009)

Mn chains on CuN



Hirjibehedin et al., Science 312, 102 (2006)

Method

Time-dependent Density Functional Theory

The time-dependent transverse magnetic susceptiblity

$$\chi^{ij}(\vec{r}t,\vec{r}'t') = \left.\frac{\delta m^i_{x,y}[B_{ext}](\vec{r}t)}{\delta B^j_{ext}(\vec{r}'t')}\right|_{B_{ext}=0,n_0}$$

can be rewritten within the atomic sphere approximation (ASA) as

$$\overline{\chi}^{ij}(rt,r't') = 4\pi\left.\frac{\delta m^i_{x,y}[B_{ext}](rt)}{\delta B^j_{ext}(r't')}\right|_{B_{ext}=0,n_0}$$

where

$$\overline{\chi}^{ij}(rt,r't') = \sum_{LL_1}\chi^{iLL_1jL_1L}$$

After repeating the same procedure for the Kohn-Sham susceptibility, one builds a Dyson equation:

$$\overline{\chi}^{ij}(r,r';\omega) = \overline{\chi}^{ij}_0(r,r';\omega) + \sum_{kl}\int dr''\int dr'''\overline{\chi}^{ik}_0(r,r'';\omega)U^{kl}(r'',r''';\omega)\overline{\chi}^{lj}(r''',r;\omega)$$

where $U^{ij}(r,r';\omega) = \frac{B^i_{eff}(r;0)}{4\pi m^i_z(r;0)}\delta_{r,r'}\delta_{i,j}$

Sum rule for Goldstone mode

The Kohn-Sham susceptibility is given by:

$$\chi^{ij}_0(\vec{r},\vec{r}';\omega) = -\frac{1}{\pi}\int dz f(z)(G^{\downarrow}_{ij}(\vec{r},\vec{r}';z+\omega)\mathrm{Im}G^{\uparrow}_{ji}(\vec{r}',\vec{r};z) + \mathrm{Im}G^{\downarrow}_{ij}(\vec{r},\vec{r}';z)G^{\uparrow}_{ji}(\vec{r}',\vec{r};z+\omega))$$

One can demonstrate that:

$$\sum_j\int d\vec{r}'\chi^{ij}_0(\vec{r},\vec{r}';\omega=0)B^j_{eff}(\vec{r}';\omega=0) = m^i_z(\vec{r};\omega=0)$$

Using the atomic sphere approximation, the previous equation can be rewritten in terms of matrix notation:

$$\sum_j\overline{\chi}^{ij}_0 B^j_{eff} = 4\pi m^i_z$$

from which *U can be extracted:*

$$U^i = \Gamma^{-1}m^i_z \quad with \quad U^i(r) = \frac{B^i_{eff}(r;0)}{4\pi m^i_z(r;0)} \quad and \quad \Gamma = \chi_0 m_z$$

U leads to the lowest eigenvalue of the denominator of the Dyson equation associated with the magnetic moments as components of the eigenvectors.

Calculate Kohn-Sham Susceptibility

It can be amazingly simplified in practice if separated into a sum of two terms *I1* and *I2*.

$$I^{ij}_1(\vec{r},\vec{r}';\omega) = \frac{i}{2\pi}\int^{E_F} dz f(z)(G^{\downarrow}_{ij}(\vec{r},\vec{r}';z+\omega)G^{\uparrow}_{ji}(\vec{r}',\vec{r};z) - G^{\downarrow*}_{ji}(\vec{r}',\vec{r};z)G^{\uparrow}_{ij}(\vec{r},\vec{r}';z-\omega))$$

$$I^{ij}_2(\vec{r},\vec{r}';\omega) = \frac{i}{2\pi}\int^{E_F} dz f(z)(-G^{\downarrow}_{ij}(\vec{r},\vec{r}';z+\omega)G^{\uparrow*}_{ij}(\vec{r},\vec{r}';z) + G^{\downarrow}_{ij}(\vec{r},\vec{r}';z)G^{\uparrow*}_{ij}(\vec{r},\vec{r}';z-\omega))$$

The first term can be computed using regular integration in the complex energy plan. The second term could be calculated along the real axis since it involves a small energy portion:

$$I^{ij}_2(\vec{r},\vec{r}';\omega) = -\frac{i}{2\pi}\int_{E_F-\omega}^{E_F} dz G^{\downarrow}_{ij}(\vec{r},\vec{r}';z+\omega)G^{\uparrow*}_{ij}(\vec{r},\vec{r}';z)$$

After doing a variable change, one could redefine other two terms whose sum leads to the Kohn-Sham susceptibility

$$\overline{I}^{ij}_1(\vec{r},\vec{r}';\omega) = \frac{i}{2\pi}\int^{E_F} dz(f(z-\omega)G^{\downarrow}_{ij}(\vec{r},\vec{r}';z)G^{\uparrow}_{ji}(\vec{r}',\vec{r};z-\omega) - f(z)G^{\downarrow*}_{ji}(\vec{r}',\vec{r};z)G^{\uparrow*}_{ij}(\vec{r},\vec{r}';z-\omega))$$

$$\overline{I}^{ij}_2(\vec{r},\vec{r}';\omega) = \frac{i}{2\pi}\int_{E_F-\omega}^{E_F} dz G^{\downarrow}_{ij}(\vec{r},\vec{r}';z+\omega)(G^{\uparrow}_{ji}(\vec{r}',\vec{r};z) - G^{\uparrow*}_{ij}(\vec{r},\vec{r}';z))$$

Evaluation of Green functions

In the Korringa-Kohn-Rostoker Green Function method, the Green function is written as:

$$G_{ij}(\vec{r},\vec{r}';z) = \sum_{LL_1} -i\sqrt{z}R^{iL}(\vec{r}_<;z)H^{iL}(\vec{r}_>;z)\delta_{ij,LL_1} + R^{iL}(\vec{r},z)G^{iL,jL_1}_B(z)R^{jL_1}(\vec{r}';z)$$

In the spectral representation

$$G_{ij}(\vec{r},\vec{r}';z) = \sum_{\vec{k}}\sum_{LL_1}\frac{\alpha^i_L(E_{\vec{k}})R^i_L(\vec{r};E_{\vec{k}})\alpha^{j*}_{L_1}(E_{\vec{k}})R^{j*}_{L_1}(\vec{r}';E_{\vec{k}})}{z-E_{\vec{k}}}$$

We could make an approximation in evaluating the wave functions *R*. One could linearize them. We choose, however, an energy independent wave function: the regular solution defined at the Fermi energy. This is a reasonable approximation for the problems of our interest.

$$G_{ij}(\vec{r},\vec{r}';z) \sim \sum_{\vec{k}}\sum_{LL_1}\frac{\beta^i_L(E_{\vec{k}})\phi^i_L(\vec{r})\beta^{j*}_{L_1}(E_{\vec{k}})\phi^{j*}_{L_1}(\vec{r}')}{z-E_{\vec{k}}}$$

or

$$G_{ij}(\vec{r},\vec{r}';z) \sim \sum_{LL_1}\phi^i_L(\vec{r})\overline{G}^{LL_1}_{ij}(z)\phi^{j*}_{L1}(\vec{r}')$$

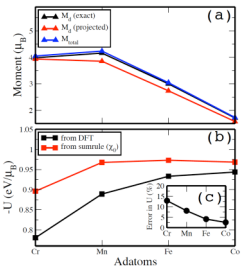
Thus

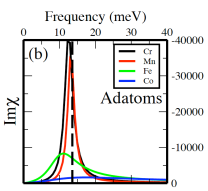
$$\overline{G}^{LL_1}_{ij}(z) = \frac{\int\int d\vec{r}d\vec{r}'\phi^{iL*}(\vec{r})G_{ij}(\vec{r},\vec{r}';z)\phi^{jL_1}(\vec{r}')}{\int dr\phi^{iL*}(r)\phi^{iL}(r)\int dr'\phi^{jL_1}(r')\phi^{jL_1*}(r')}$$

Finally

$$\overline{G}^{LL_1}_{ij}(z) = \sum_{L_2L_3}\Big(-i\sqrt{z}\int_0^{r_{ws}}d\vec{r}H^{iL_2}(\vec{r}';z)\psi^{iL}(\vec{r}')\int_0^{r'}d\vec{r}\psi^{iL_1*}(\vec{r})R^{iL_2}(\vec{r},z)\delta_{ij,L_2L_3} - i\sqrt{z}\int_0^{r_{ws}}d\vec{r}R^{iL}_2(\vec{r}',z)\psi^{iL}(\vec{r}')\int_{r'}^{r_{ws}}d\vec{r}\psi^{jL_1*}(\vec{r})H^{jL_2}(\vec{r},z)\delta_{ij,L_2L_3} + \int_0^{r_{ws}}d\vec{r}\psi^{jL*}(\vec{r})R^{jL_2}(\vec{r},z)G^{jL_2,jL_3}_B(z)\int_0^{r_{ws}}d\vec{r}R^{jL_3}(\vec{r}';z)\psi^{jL_1}(\vec{r}')\Big)$$

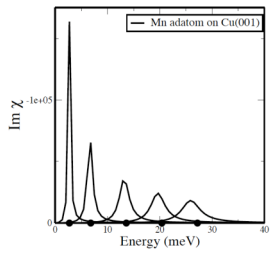
Adatoms





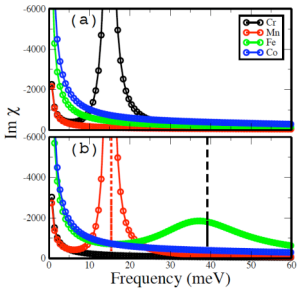
From the Heisenberg model, a delta function is expected at the Larmor Frequency given by *gB*.

Adatoms



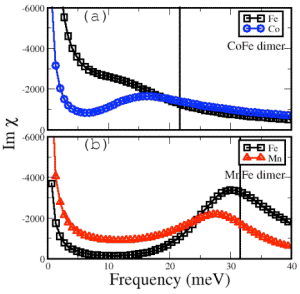
Damping increases by increasing the magnitude of the field

Dimers made of similar adatoms



From the Heisenberg model, optical mode is expected at 2J(M1-M2)/M1M2, J being the magnetic exchange interaction

Dimers made of dissimilar adatoms



Conclusion

We have shown that a simple approach, based on TD-DFT and the KKR-GF method, can be used to extract dynamics magnetic susceptibilities. The description of the electronic structure is embedded into our single particle Green functions. The size of matrices in the Dyson equation are small enough to permit calculations of large nanostructures in future applications. We have developed an identity that leads to a numerically stable method of extracting the proper Coulomb interaction to be used in the Dyson equation from which the full dynamical susceptibility is obtained. As an application, 3*d* adatoms and dimers deposited on Cu(001) surface were investigated from first-principles.

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