Surface imaging, inelastic scattering, and STM measurements

J. Fransson
Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA and
Center for Nonlinear Studies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

Surface imaging of inelastic Friedel oscillations [1] — LA-UR-06-7425

Inelastic electron tunneling spectroscopy (IETS) with scanning tunneling microscope, IETS-STM, is a well-established technique, starting with an important experiment by Stipe *et al.* [2]. These experiments show step like features in the tunneling current and local density of states (DOS). The physical explanation of the effect is straightforward: once the energy of the tunneling electron exceeds the energy required to excite local vibrations, there is a new scattering process that contributes to the scattering of electrons due to inelastic excitation of the local mode [3]. The local vibrational modes can be seen in differential conductance measurements as sidebands to the main (elastic) conductance peak [4]. Recently, IETS-STM was used to measure the spin excitations of individual magnetic atoms [5]. In addition, recent experiments show dominant inelastic channels which are strongly spatially localized to particular regions of a molecule [6].

We propose [1] to use resolution of STM to address spatially resolved inelastic tunneling features produced by local inelastic scattering of the molecule both near the scattering center and at far distances. The "fingerprint" of the inelastic scattering will be present even away from the scattering center. Using local IETS-STM would enable measurements of the standing waves produced by inelastic scattering off impurities, which could be revealed as waves seen in the *second derivative* $\partial^2 I(\mathbf{r},V)/\partial V^2$. For the simplest model of parabolic conduction band these waves will be seen as standing waves with period set by the Fermi momentum k_F. These standing waves are seen in the oscillations of the $\partial^2 I(\mathbf{r},V)/\partial V^2$ and are *similar but qualitatively different* from the conventional Friedel oscillations.

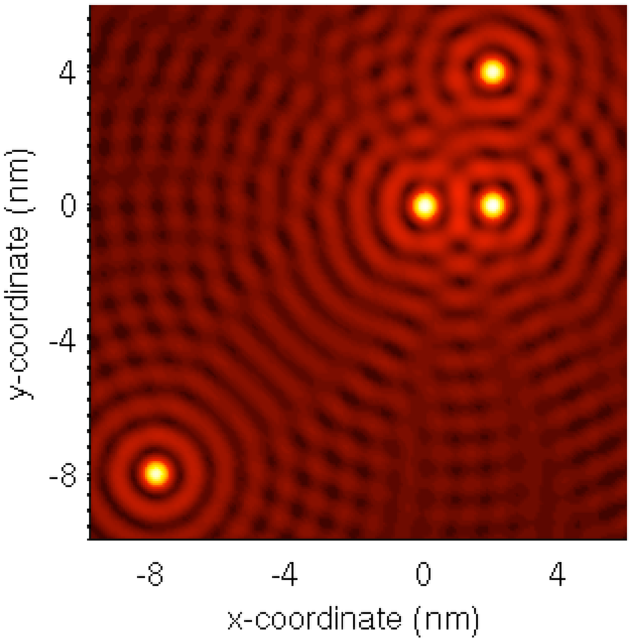

FIG. 1: Qualitatively expected IEST-STM resonance, e.g. at phonon mode resonance, signal on surface with four inelastic impurities embedded. Here we used $\omega_0 = 5$ meV, Fermi energy $E_F \simeq 0.45$ eV, and $T = 10$ K.

In case of regular Friedel oscillations the charge that screens off the impurity exhibit oscillations at large distances from the impurity [7]. Recent STM measurements have observed standing wave patterns in the elastic scattering channels on surfaces with atomic impurities adsorbed on the surface [8, 9], sometimes referred to as energy resolved Friedel oscillations and is the modulated local density of states (DOS) formed by scattering and interference in the surface electron gas. These oscillations are seen in a wide range of bias as they reflect screening of the charge by electron states in the whole bandwidth. Inelastic scattering of surface electrons off the molecules may be viewed as *inelastic* Friedel oscillations produced by the electron states that are involved in screening. Inelastic Friedel oscillations are seen only in the narrow window of energies near the energy of the mode ω_0 at which inelastic scattering occurs, in contrast to conventional Friedel screening.

Exchange interaction and Fano resonances in diatomic molecular systems [10] — LA-UR-06-6599

There are various techniques that allows one to detect and manipulate spin states in the solid state, which attract lots of interest. A partial list include optical detection of electron spin resonance (ESR) in a single molecule [11], tunneling through a quantum dot [12], and, more recently, ESR-scanning tunneling microscopy (ESR-STM) technique [13, 14]. Experimentally, modulation in the tunneling current has been observed by STM using spin-unpolarized electron beam [12, 13] .

Typically in STM measurements with an object located on a substrate surface, the tunneling current can either go directly between the STM tip and the substrate or go via the object. The tunneling electrons are thus branched between different pathways, which gives rise to

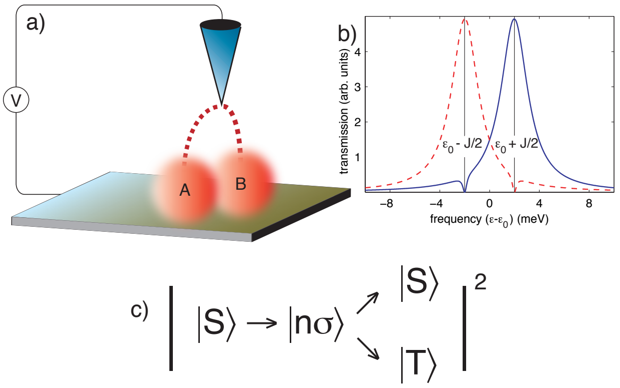

FIG. 2: a) Diagram of the diatomic $A+B$ molecule coupled to the tip and substrate. The particles interact via exchange interaction J. b) Transmission for transitions between the singlet/triplet and the anti-bonding (dashed) and bonding (solid) one-electron states. c) Phase space branching of the tunneling wave functions between the singlet $|S\rangle$ and triplet $|T\rangle$ states. The respective amplitudes of the tunneling through these states have to be added, resulting in the Fano like features.

interference effects when the partial waves merge into one in the tip or the substrate [15]. This interference leads to a suppressed transmission probability for the tunneling electrons at certain energies. The suppressed transmission is a fingerprint of Fano resonances [16], and generally appear in systems where tunneling electrons are branched between different pathways.

We propose a method to measure the two-electron singlet-triplet (S-T) exchange splitting J in a diatomic molecule by means of STM [10]. The presence of two pathways for the tunneling current between the tip and the substrate, through the diatomic molecule, gives rise to interference effects (Fano resonance) between the electron waves traveling through the singlet and triplet states. In the direct tunneling between the tip and substrate via the molecule, the probability for the tunneling is proportional to Γ_0. In addition, because of the phase space branching of tunneling possibilities, the tunneling probability is multiplied with the interference probabilitiy γ^2, hence, the characteristic energy width of the anti-resonance is $\gamma^2\Gamma_0$. Clearly, the anti-resonances will be measurable in the second derivative of the current whenever $\gamma \ll 1$. Such measurements will be extremely useful in situations where the level broadening is larger than the singlet-triplet splitting, since a large broadening smears all features and therefore prevent identification of those states in the differential conductance.

Fano resonances can be realized in a variety of system, ranging from systems with interactions between continuum states and a localized state, to systems where the branching of the wave function through diatomic molecules. In the case we consider here, we also have to include the fact that the one-electron states in a two-level system with the levels being resonant, consist of an anti-bonding and a bonding state (both being spin-degenerate). This modifies the expected transport properties such that transitions between the triplet and the anti-bonding one-electron state generate a dip in the transmission at the energy for this transition, see Fig. 2 b) (dashed). On the other hand, transitions between the singlet and the bonding one-electron state give rise to a dip in the transmission at the energy for this transition, see Fig. 2 b) (solid). In turn, the features that appear at voltages corresponding to both the singlet and triplet states, provide a unique fingerprint which allows the read out of the S-T exchange splitting J.

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[1] J. Fransson and A. V. Balatsky, cond-mat/0701606.
[2] B. C. Stipe, M. A. Rezaei, and W. Ho, Science, **280**, 1732 (1998).
[3] R. C. Jaklevich and J. Lambe, Phys. Rev. Lett. **17**, 1139 (1966); J. Lambe and R. C. Jaklevich, Phys. Rev. **165**, 821 (1968).
[4] H. Park, J. Park, A. K. L. Lim, E. H. Anderson, A. P. Alivisatos, and P. L. McEuen, Nature, **407**, 57 (2000).
[5] A. J. Heinrich, J. A. Gupta, C. P. Lutz, and D. M. Eigler, Science, **306**, 466 (2004); C. F. Hirjibehedin, C. P. Lutz, and A. J. Heinrich, Science, **312**, 1021 (2006).
[6] M. Grobis, K. H. Khoo, R. Yamachika, X. Lu, K. Nagaoka, S. G. Louie, M. F. Crommie, H. Kato, and H. Shinohara, Phys. Rev. Lett. **94**, 136802 (2005).
[7] J. Friedel, Nuovo Cimento Suppl. **7**, 287 (1958).
[8] Y. Hasegawa and Ph. Avouris, Phys. Rev. Lett. **71**, 1071 (1993).
[9] M. F. Crommie, C. P. Lutz, and D. M. Eigler, Nature, **363**, 524 (1993).
[10] J. Fransson and A. V. Balatsky, Phys. Rev. B (Brief Reports) *in press* (2007); cond-mat/0609324.
[11] J. Köhler, J. A. J. M. Disselhorst, M. C. J. M. Donckers, E. J. J. Groenen, J. Schmidt, and W. E. Moerner, Nature **363**, 242 (1993); J. Wrachtrup, C. von Borczyskowski, J. Bernard, M. Orrit, and R. Brown, *ibid.* **363**, 244 (1993); Phys. Rev. Lett. **71**, 3565 (1993).
[12] H. -A. Engel and D. Loss, Phys. Rev. Lett. **86**, 4648 (2001); Phys. Rev. B, **65**, 195321 (2002).
[13] Y. Manassen, R. J. Hamers, J. E. Demuth, and A. J. Castellano, Jr., Phys. Rev. Lett. **62**, 2531 (1989); D. Shachal and Y. Manassen, Phys. Rev. B, **46**, 4795 (1992); Y. Manassen, J. Magn. Reson. **126**, 133, (1997); Y. Manassen, I. Mukhopadhyay, and N. R. Rao, Phys. Rev. B, **61**, 16223 (2000).
[14] C. Durkan and M. E. Welland, Apply. Phys. Lett. **80**, 458 (2002).
[15] V. Madhavan, W. Chen, T. Jamneala, M. F. Crommie, and N. S. Wingreen, Phys. Rev. B, **64**, 165412 (2001).
[16] U. Fano, Phys. Rev. **124**, 1866 (1961).