Tunneling Experiments on Manganites

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Abstract

Tunnel investigations of ceramic and film $La_{0.57}Ca_{0.43}MnO_3$ (LCMO) compounds are carried out. Besides the well known from Raman spectra phonon features up to 80 meV, the presence of bosonic excitations in the region above 100 meV has been obtained. Qualitative changes of tunneling spectra (below and above 100 meV) caused by chemical modification of the oxide surface after application of high voltages to the junction were revealed. Results are discussed within a scenario involving tunnel magnetoresistance (TMR) and the possible manifestation of orbitons, novel excitation recently predicted in manganites.

Key words: manganites; excitation spectra; tunneling measurements; orbitons

Manganite compounds are strong-correlated systems involving charge interactions with orbital degrees of freedom besides standard - magnetic and lattice ones. Orbital order creates novel excitations - orbitons with energies above those of magnons and phonons (up to 80 meV) [1]. Our study is focused on tunneling spectroscopy of high-energy excitations in the LCMO manganites. The main goals are to improve the understanding of the electron-boson interaction role in manganite properties and to elucidate the TMR nature. Experimental approach and analysis of tunnel data for normal-state metal-insulator-metal (N-I-N) contacts are based on the concept proposed in the paper [2]. Even (odd) $\sigma_{\pm}(V) = (\sigma(+V) \pm \sigma(-V))/2$ parts of the differential conductance $\sigma(V) = dI/dV$ have been discriminated. The function of the electron-boson interaction in a bulk electrodes $g(\omega)$ is proportional to $\int \frac{d\sigma_{-}(V)}{dV} \frac{dV}{V^2-(\omega/e)^2}$, whereas $d\sigma_{+}(V)/dV$ characterizes the density of bosonic excitations in the insulating barrier.

The results reported here have been obtained for the bulk system $La_{0.57}Ca_{0.43}MnO_3$ close to charge orbital ordering phase and, as we assume, is mostly convenient for a possible orbiton observation. The contacts with a silver counterelectrode were formed by vacuum evaporation of films or pressing a sharp Ag tip into the manganite surface. The junction resistances at 77 K were about several hundreds of Ohms showing that we are dealing with a potential barrier between two metallic electrodes. The barrier is formed by a degraded layer on the manganite surface that appears as a result of oxygen losses in the near-interface region.

Tunneling processes through the degraded region causes an anomalous conductance-versus-voltage dependence $\sigma(V) = a\sqrt{V} + b \mid V \mid$ that is a common feature of manganites and cuprates [3]. The statement is supported by our data presented in fig. 1a and can be explained in terms of the electron-electron interaction and inelastic tunneling across magnetically active localization states in the barrier. The derivative $d\sigma_{+}(V)/dV$ shows a broad continuum of excitations in the barrier with a nonprominent structure reflecting details of that are more visible for voltages below

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100 meV where magnon and phonon excitations are expected. In our opinion, electron scatterings on the broad continuum of magnetic-like excitations is the reason of the TMR drop with increasing the voltage clearly seen in the inset in fig. 1b.

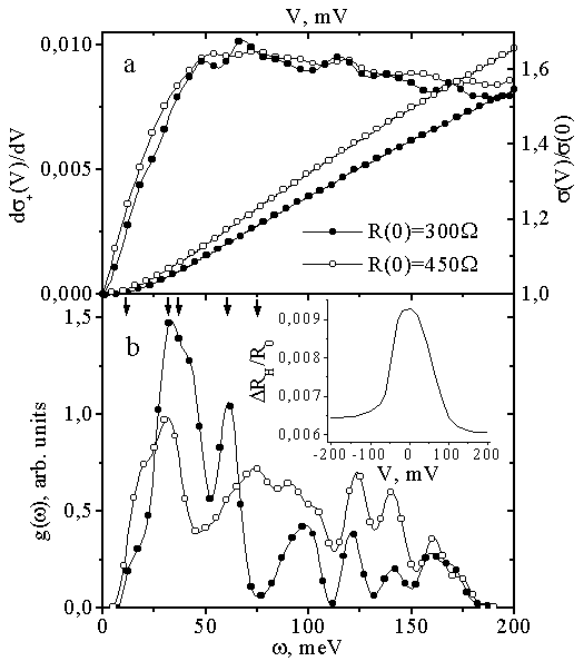

Fig. 1. Tunneling data for Ag - I - LCMO. (a) The upper panel shows the even part of the differential conductance normalized to its zero-bias value (the right axis) and its derivative over voltage for an initial junction with the zero-voltage resistance of 450 Ohm (T_c=180 K) (open circles), and the same junction with the 300 Ohm (T_c=200 K) resistance after the high-voltage treatment (black circles). (b) The lower panel demonstrates the electron-boson interaction spectra for both cases and the normalized zero-bias tunnel magnetoresistance for an initial junction (the inset). The arrows indicate positions of phonon features as they were estimated in Raman scattering experiments [4,5]

Consider the bulk excitation spectrum that was reconstructed from the odd part of $\sigma_-(V)$ (see fig. 1b). There are prominent peaks close to phonon modes of the material known from Raman data (they are indicated by arrows in fig. 1b), as well as from far-infrared absorption and inelastic neutron scattering data, the same time there is a structure above 80 meV that cannot be related from energy reasons to standard magnon or phonon scattering processes. Part of these high energy peaks which manifests obvious multiplication by energy of previous ones could be interpreted as two- and three-phonon features. Their appearance is expected for systems with lattice instability like Jahn-Teller distortions. Increase of the conductivity leads to a suppression of distortions (a system becomes more cubic) and to lower anharmonicity.

To explain a peak at 95 mV, we propose to account for orbital fluctuations which create a new channel of carrier inelastic scatterings in the underdoped regime. The maximal loss of energy could be estimated as an energy of orbitons in an orbitally ordered insulating state. Energies of the novel excitations are nearly those of two-phonon processes. Numerous Raman studies show some softening and widening of the features under doping increase [4].

To give an additional argument in support of the last supposition, we applied to the junction high voltage biases that cause in perovskites strong chemical modifications of the interface (supposedly, oxygen ion displacements). As it was shown in our paper [3], a radical (but reversible) effect of oxygen electromigration processes results in reducing the junction resistance but - what is more important - its effect is the enrichment of upper layers by oxygen. It has obtained that the surface T_c changed from 180 K for the initial tunnel resistance 450 Ohm at $V = 0$ to 200 K for 300 Ohm after application of high biases up to 1 V. A simultaneous increase of the electron-boson interaction should be noticed (see the amplitude changes of $g(\omega)$ in fig. 1b). When a metallization of the interface is increased by the electromigration processes, the amplitude of the structure above 80 mV decreases while the low-energy structure is enhanced. It should be emphasized that the maximum which disappears at nearly 75 mV is known as a breathing mode. Such a suppression is well known in Raman spectra [5]. In spite of a clear decrease of multi-phonon features, the intensity of a feature at 95 mV has not been changed radically.

Resuming, we have performed tunneling measurements of high-energy features of manganites and found an unusual structure in the voltage region between 80 and 180 mV that decreases when an external voltage perturbation is applied to the material. Our experiments have shown that the normal-state tunneling spectroscopy can be a powerful tool for studying a novel scattering channel in manganites.

References

[1] E. Saitoh, S. Okamoto, K. T. Takahashi, K. Tobe, K. Yamamoto, T. Kimura, S. Ishihara, S. Maekawa, Y. Tokura, Nature **410** (2001) 180.

[2] V. M. Svistunov, M. A. Belogolovskii, O. I. Chernyak, A. I. Khachaturov, A. P. Kvachev, Sov. Phys. JETP **57** (1983) 1038.

[3] M. A. Belogolovskii, Yu. F. Revenko, A. Yu. Gerasimenko, V. M. Svistunov, E. Hatta, G. Plitnik, V. E. Shaternik, E. M. Rudenko, Fiz. Nizkikh Temp. **28** (2002) 553.

[4] V. A. Amelitchev, B. Guttler, O. Yu. Gorbenko, A. R. Kaul, A. A. Bosak, A. Yu. Ganin, Phys. Rev. B **63** (2001) 104430.

[5] E. Liarokapis, Th. Leventori, D. Lampakis, D. Palles, J. J. Neumeier, D. H. Goodwin, Phys. Rev. B **60** (1999) 12758.