## Manifestation of multiphonon structure and orbitons in the tunnel spectra of manganites

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The study results of the tunnel conductance of heterostructures formed by a silver tip and a doped  $Ag/La_{0.57}Ca_{0.43}MnO_3$  manganite in the voltage region above manganite phonon and magnon frequencies are presented. Three maxima have been observed at energies corresponding to the peaks in the Raman spectra of  $LaMnO_3$ . It is supposed that the maxima are due to the interaction of tunneling electrons with excitations in the orbital subsystem of a degraded region nearby the metal-manganite interface.

Представлены результаты исследований туннельной проводимости гетероструктур, образованных серебряным острием и легированным манганитом Ag/La<sub>0,57</sub>Ca<sub>0,43</sub>MnO<sub>3</sub>, в области напряжений выше фононных и магнонных частот манганитов. Обнаружены три максимума при энергиях, соответствующих пикам в спектрах комбинационного рассеяния LaMnO<sub>3</sub>. Предполагается, что указанные максимумы обусловлены взаимодействием туннелирующих электронов с возбуждениями в орбитальной подсистеме деградированной области, примыкающей к интерфейсу металл — манганит.

The interest arised to ferromagnetic heterostructures based on La<sub>1-x</sub>Ca<sub>x</sub>MnO<sub>3</sub> type manganites due to discovery of Giant magnetoresistance (GMR) in such systems. From the fundamental and practical point of view, the GMR dependence on the voltage applied to the tunnel contact is of interest. The scattering on magnetic excitations in a tunnel barrier which invert a spin and thereby inhibit the effect is supposed to be possible mechanism responsible for this effect. Actually, to substantiate the assumption about the connection of the voltage GMS dependence with intensity of inelastic processes in the barrier, it is necessary to reveal and investigate the presence of excitations with high energies (above 100 meV) inside the barrier or in a near-barrier region. It is assumed that the scattering on phonons, and other quasi-particles (magnons, etc.) may play noticeable part in the GMR suppression, besides magnetic processes.

Strongly correlated systems, such as manganites possessing colossal magnetoresistance and high temperature superconductors, are promising from the viewpoint of orbital wave observation, because they contain the transitional metal ions for which the orbital degree of freedom is of importance [2, 3]. The orbitally ordered states were discovered in a some transitional metal compounds [4, 5], and orbitons was theoretically predicted for LaMnO<sub>3</sub> [6, 7]. In this work, the tunnel spectrums of bulk La<sub>0.57</sub>Ca<sub>0.43</sub>MnO<sub>3</sub> (LCMO) polycrystals were studied.

The experimental LCMO samples have been obtained by standard solid-phase synthesis from the initial rare-earth element,

calcium and manganese oxides at the synthesis temperature  $T_s = 1200-1250$ °C in air with the repeated grinding, pressing, and annealing. As a potential barrier to the tunneling electrons, the natural oxide layer arising on the manganite surface was used [8]. Oxidation was conducted at the natural cooling in a laboratory furnace down to room temperature. The metallic injector in the studied manganite heterostructures was a pointed silver wire pressed against the sample with a force that could be regulated by an adjusting mechanism. Measuring the first derivative of current-vs-voltage characteristics I(V) of the tunnel contacts on voltage  $\sigma(V) = dI(V)/dV$  was carried out by a standard modulation method in the preset voltage mode.

The tunnel spectroscopy of bosonic excitations in non-superconductive metals [9] is based on the fact that odd part of differential conductivity  $\sigma_{-}(V) = (\sigma(+V) - \sigma(-V))/2$  of a metal-insulator-metal tunnel contact at temperatures near to zero is in proportion to real part  $\sigma(\omega)$  of electronic excitations in the conductor. It is supposed that the interaction of electrons with bosons in other metallic plate can be neglected:

$$\sigma(V) = -\alpha \sigma_0 \operatorname{Re}\sigma(eV) / \varepsilon_F. \tag{1}$$

Here  $\sigma_0 = \sigma(V = 0)$  is the conductivity in zero voltage;  $\varepsilon_F$ , the Fermi energy, dimensionless constant  $\alpha \approx 1$  [9]. The physical nature of formula (1) is defined by two factors: first, by the dependence of dielectric layer transparency in MIM transition on the energy  $\omega$  of electron tunneling therethrough [9] and second, by the dependence of electronic state density in the conductor under investigation on  $\omega$  [10]. While in ordinary metals, the main contribution to  $\sigma(V)$  is related to the first factor [9], in the case of manganites, dominant is the energy-dependent variation of their electronic characteristics due to the relatively low values of Fermi energies  $\epsilon_F$ . The estimation of  $\epsilon_F$  in the framework of free electron gas model gives for alloyed manganites, showing the colossal magnetoresistance effect, a value not exceeding few hundreds meV [11, 12]. Formula (1) allows to calculate from experimental  $\sigma$  (V) dependence the imaginary part  $\sigma(\omega)$ , and then to determine the electronboson interaction function (see [9, 10]):

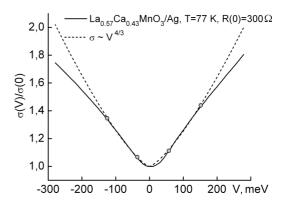


Fig. 1. Barrier characteristics of Ag heterocontact with a  $\text{La}_{0.57}\text{Ca}_{0.43}\text{MnO}_3$  film sample. The hetero-contact resistance  $R(V=0)=R_0$  is 300  $\Omega$ , the measurement temperature 77 K. The points indicate the voltage ranges where the theoretical amd experimental data are in agreement.

$$g(\omega) = \frac{2\omega\varepsilon_F}{\alpha\sigma_0\pi^2} \int_0^\infty \frac{d\sigma_-(V)}{dV} \frac{dV}{\omega^2 - (eV)^2}.$$
 (2)

Formula (2) makes it possible to reconstruct the electron-boson interaction functions  $g(\omega)$  for the studied objects and to calculate the values of the corresponding constants  $\lambda = 2 \int g(\omega) d\omega/\omega$ .

It has been shown [13] that in tunnel contacts, the electron transfer is carried out by elastic under-barrier tunneling through the localized states inside the barrier; this results in the power nonlinearity of amorphous layer volt-ampere characteristics:  $I(V) \sim V^{7/3}$ , and the tunnel conductivity variation is described as  $\sigma(V) \sim V^{4/3}$  [14]. For the studied Ag-La<sub>0.57</sub>Ca<sub>0.43</sub>MnO<sub>3</sub> system, an agreement with the theory is observed in a wide region of self-energy excitations and extrinsic contributions (up to 130 meV) (Fig. 1).

For the system  $La_{0.57}Ca_{0.43}MnO_3$ , the reconstructed spectral characteristics of electron-phonon interaction  $g(\omega)$  are presented in Fig. 1. The area of self-energy excitations (which reflects the tunnel barrier properties) spreads up to 100~meV. The most clearly expressed singularities of this area are demonstrated in the spectra of phonons and magnons. Comparison of the singularity sites in the phonon and magnon spectra shows a good agreement with the

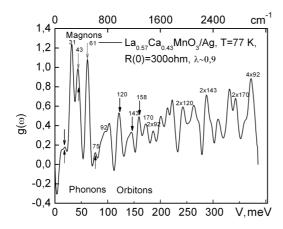


Fig. 2. The electron-bozon interaction function  $g(\omega)$  for Ag hetero-contact with a  $La_{0.57}Ca_{0.43}MnO_3$  bulk sample derived from the odd part of differential conductance. The sites of main peaks in the Raman spectrum of  $LaMnO_3$  are shown by arrows [1, 15, 16]. The hetero-contact resistance  $R(V=0)=R_0$  is 300  $\Omega$ , the measurement temperature 77 K.

results of optical experiments for  $La_{0.7}Ca_{0.3}MnO_3$  [15, 16].

In [1], the features of LaMnO<sub>3</sub> optical spectra were discussed in the 120-170 meV range, the peaks in this area being supposed to be connected with the display of orbital excitations. The voltage positions of three basic peaks in this interval of a tunnel spectrum (see Fig. 2) agree very well with similar three singularities in the Raman spectrum of LaMnO<sub>3</sub> with energies 125, 145 and 160 meV [1] and, accordingly, can be explained by interaction of tunneling electrons with excitations in the orbital subsystem. As for maxima at energies exceeding 160 meV, those are probably the phonon satellites of the orbital excitation area [16]. In our opinion, the above-mentioned features hardly can be explained by multiphonon processes, since even in tunnel descriptions of such strong-constrained superconductors as lead, the latter show themselves as a rather weak nonlinearity [18]. Finally, if a very strong electron-phonon interaction with a certain mode in the oscillation spectrum of lattice exists in the investigated manganites (that is possible in principle), then the observed features would be the harmonics of the same frequency, that is not observed in our experimental curves.

To conclude, it is to note that the orbital strain waves observation in the orbitally-ordered electron subsystem of manganites using the effect of electron tunneling opens new possibilities for research of this phenomenon under varying external conditions, for example, magnetic field, that will allow to investigate the orbiton interaction with other quasi-particle excitations in strongly correlated electronic environments.

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## Прояв мультифононної структури і орбітонів у тунельних спектрах манганітів

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Представлено результати досліджень тунельної провідності гетероструктур, утворених срібним вістрям і легованим манганітом Ag/La<sub>0.57</sub>Ca<sub>0.43</sub>MnO<sub>3</sub>, в області напруг вище фононних і магнонних частот манганітів. Знайдено три максимуми при енергіях, відповідних пікам у спектрах комбінаційного розсіяння LaMnO<sub>3</sub>. Передбачається, що вказані максимуми обумовлені взаємодією тунелюючих електронів із збудженнями в орбітальній підсистемі деградованої області, що примикає до інтерфейсу метал — манганіт.