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Evolution of the inter-layer coupling in bilayered manganites revealed by ferromagnetic resonance spectra

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Abstract

Ferromagnetic resonance has been used to investigate the inter-layer coupling in a bilayered manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.38$) single crystal. The coexistence of a ferromagnetic resonance line and a paramagnetic resonance line was observed over a broad temperature range. Antiferromagnetic coupling between the adjacent MnO_2 bilayers can be identified from the observation of an optical mode in the resonance spectra. Analysis of the temperature dependence of the resonance field and intensity reveals the evolution of the inter-layer coupling as a function of temperature. Our study suggests that ferromagnetic resonance provides a useful method for investigating the inter-layer coupling in bilayered manganites.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The inter-layer exchange coupling (IEC) between two ferromagnetic (FM) layers separated by a nonmagnetic spacer layer has received considerable attention due to its relevance to both the giant magnetoresistance (GMR) [1, 2] and tunneling magnetoresistance (TMR) effects [3, 4]. In most cases, the IEC is studied in artificial magnetic multilayers and tunneling junctions. In contrast, the bilayered manganites with a formula of $La_{2-2x}Sr_{1+2x}Mn_2O_7$, which have been well known for the colossal magnetoresistance (CMR) effect, provide a natural system in which to study the mechanism of IEC [5] as well as TMR [6]. The bilayered crystal is composed of MnO_2 bilayers which are stacked along the *c* axis and separated by insulating nonmagnetic $(La, Sr)_2O_2$ layers [7], as seen in the inset of figure 1. In such a quasi-two-dimensional system, the inter-bilayer as well as the intra-bilayer spin and charge dynamics are expected to critically depend on the inter-layer exchange coupling between the MnO_2 bilayers. Therefore, a full mapping of the evolution of IEC with temperature is critical to understanding the physics in bilayered manganites.

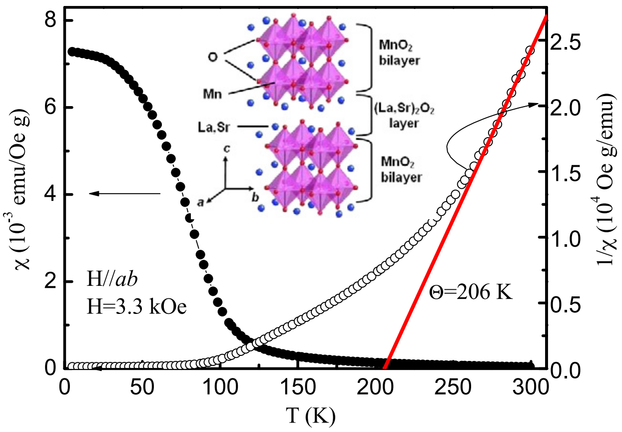

Figure 1. Temperature dependence of the dc magnetic susceptibility and inverse dc magnetic susceptibility in the *ab* plane. The inset shows the crystal structure of $La_{2-2x}Sr_{1+2x}Mn_2O_7$.

Among various techniques used for the study of IEC in magnetic multilayers, ferromagnetic resonance (FMR) has been proved to be a useful method for evaluating the IEC

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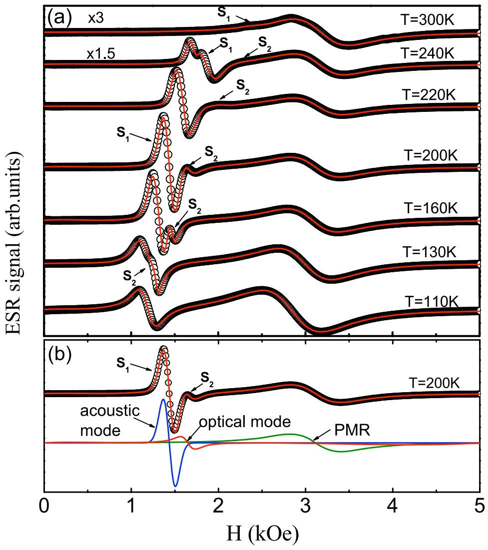

Figure 2. (a) In-plane ESR spectra of the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.38$) single crystal. (b) The measured resonance curve was resolved into several components: the FMR acoustic mode one, the FMR optical mode one, and the PMR signal. The open circles are experimental data and the solid lines are the fitted curves based on the resolved components. Signals S_1 and S_2 relate to the acoustic mode and antiparallel optical mode, respectively.

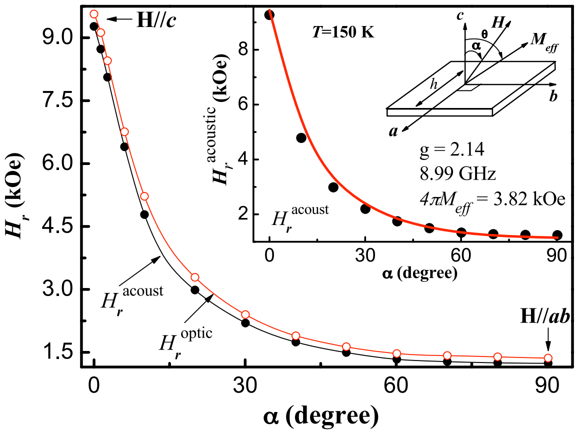

Figure 3. Angular dependence of the resonance fields H_r^{acoust} and H_r^{optic} measured at 150 K. The inset shows that the angular dependence of the resonance fields of the acoustic mode agrees well with the theoretical fit (the solid line) obtained using equations (1) and (2). The measurement configuration and the parameters obtained from the fitting are also given.

for both parallel and antiparallel coupled systems [8–10]. Theoretical study of FMR in magnetic multilayers with coupled layers gives a prediction for the behavior of acoustic and optical resonance modes occurring under the IEC effects [11, 12]. If the IEC is ferromagnetic (FM), the optical mode in the in-plane spectra appears at lower magnetic field than the acoustic mode. In contrast, if the IEC is antiferromagnetic (AFM) the optical peak appears at higher field than the acoustic peak. If the two FM layers are exactly identical, the optical mode cannot be observed in the FMR spectra. The mode positions and intensities depend on the exchange field and strength of the inter-layer coupling. As a result, the separation of resonance fields between the optical and acoustic modes and the resonance intensity ratio can be used to estimate the relative IEC strength.

In this work, we have investigated the temperature dependence of the IEC in a bilayered manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.38$) single crystal by means of FMR. To the best of our knowledge, this is the first report of studying inter-layer coupling in bilayered manganites by using FMR. The analysis of the FMR spectra reveals the presence of AFM inter-layer coupling above the Curie temperature T_C and its evolution with temperature, which is consistent with previous neutron scattering results. Our study suggests that FMR provides a useful method for investigating the inter-layer coupling in bilayered manganites.

2. Experimental details

Single crystals of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.38$) were grown by the floating-zone method in an optical image furnace. X-ray diffraction measurement on the powders shows that the crystal grown was a single phase without any impurity phase. It has a tetragonal symmetry with a space group $I4/mmm$ ($a = 0.387\,142(8)$ nm, $c = 2.019\,17(4)$ nm) which is consistent with the previous reports [13]. The crystal was oriented using Laue x-ray diffraction patterns, and cut into a rectangular shape with a size of $1 \times 1 \times 0.1$ mm^3. The largest plane is the easy plane (ab plane) with the c axis normal to the ab plane. The dc susceptibility was measured using a superconducting quantum interference device magnetometer (Quantum Design, MPMS-7). The FMR experiments were carried out using a JEOL JES-FA200 ESR spectrometer at X-band frequencies ($\nu \approx 9.4$ GHz) in the temperature range from 100 to 300 K. The in-plane spectra were measured with both the microwave field h and applied field H parallel to the ab plane. The angular dependence of the resonance was measured by rotating the sample from $H \parallel c$ axis to $H \parallel ab$ plane. The configuration of the measurement was shown in the inset of figure 3.

3. Results and discussion

Figure 1 shows the temperature dependence of the dc magnetic susceptibility with field applied along the ab plane. At high temperature, the susceptibility increases slowly with decreasing temperature and the inverse magnetic susceptibility $1/\chi$ exhibits a linear dependence with temperature. The slope of $1/\chi$ changes, with departure from the Curie–Weiss law at about 210 K, below which in-plane 2D FM correlations are established in the MnO_2 layers. The magnetization starts to increase rapidly at 110 K, which corresponds to the 3D FM ordering temperature T_C.

Figure 2 shows the in-plane FMR spectra of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.38$) at selected temperatures from 100 to 300 K. Above the Curie temperature $T_C \sim 110$ K, the spectra consist of a paramagnetic resonance (PMR) line and several FMR signals. The coexistence of PMR and FMR signals within a broad temperature range has been widely observed in manganites and indicates the intrinsic phase

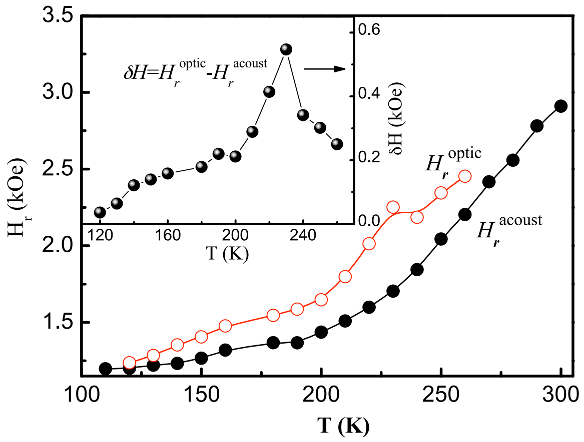

Figure 4. Temperature dependence of the resonance field for the acoustic and antiparallel optical modes. The inset shows the temperature dependence of the field separation $\delta H = H_r^{\mathrm{optic}} - H_r^{\mathrm{acoust}}$.

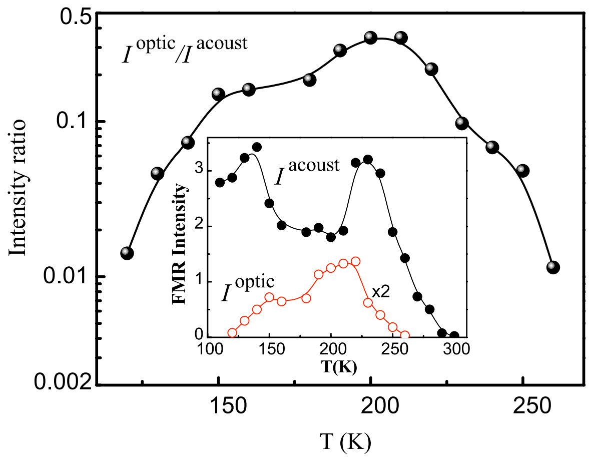

Figure 5. Temperature dependence of the intensity ratio $I^{\mathrm{optic}}/I^{\mathrm{acoust}}$ between the acoustic and antiparallel optical modes. The inset shows the intensity for the two resonance modes.

separation in the system [14, 15]. In this study, we focus on the FMR signals, which provide information on both the intra-layer and inter-layer spin couplings.

The FMR signal (S_1) appears on the lower field side of the PMR line when the temperature is below 300 K. This signal grows with decreasing temperature and can be assigned to the acoustic mode. The presence of the acoustic mode in the paramagnetic state should be attributed to the emergence of local FM correlations within MnO_2 bilayers [16]. The acoustic mode signal shifts to lower field and separates into multiple peaks with decreasing temperature. This multi-peak feature for the FMR acoustic mode could be due to the inhomogeneous distribution of different FM clusters in the MnO_2 layers. When the temperature is lower than 220 K, at which the in-plane 2D FM correlation forms, all the peaks of the acoustic mode merge into one main peak. At about 260 K, another FMR signal (S_2) appears on the higher field side of the main acoustic mode S_1 and can be assigned to the antiparallel optical mode, which implies the appearance of local AFM coupling across MnO_2 bilayers. As temperature decreases, the optical mode signal approaches the acoustic one and its intensity initially increases and then decreases slowly below 220 K, as shown in figures 2 and 3. At about 110 K, the optical peak of S_2 merges into the acoustic peak of S_1 and cannot be observed in the spectra. The disappearance of the AFM optical mode is attributed to the formation of 3D FM ordering below $T_C \sim 110$ K. This observation of antiparallel optical modes is consistent with the neutron scattering studies on $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.4$) [17] which showed that the AFM correlations coexist with FM critical fluctuations in the paramagnetic state and disappear rapidly as the system enters into the 3D FM state on cooling. In order to obtain the resonance parameters, each resonance curve is resolved into several components which give the best fit to experimental data, as shown in figure 2(b). Both the resonance fields and intensities for the acoustic and antiparallel optical modes are obtained through the fits of the spectra.

The angular dependence of the resonance fields H_r measured at 150 K is shown in figure 3. It is clear that the optical mode is at higher field than the acoustic mode for all directions, indicating an antiferromagnetic coupling between adjacent MnO_2 bilayers. The angular dependence of the resonance fields has been fitted to the Kittel equation [18]:

$$\left(\frac{\omega}{\gamma}\right)^2 = (H\cos\alpha - 4\pi M_{\mathrm{eff}}\cos\theta)^2 + H\sin\alpha(H\sin\alpha + 4\pi M_{\mathrm{eff}}\sin\theta), \quad (1)$$

where $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio, $4\pi M_{\mathrm{eff}}$ is the effective magnetization, α is the angle of H with respect to the film normal, θ is the inclination of the magnetization from the film normal and given by

$$\frac{\sin(\theta-\alpha)}{\sin\theta\cos\theta} = \frac{4\pi M_{\mathrm{eff}}}{H}. \quad (2)$$

The line obtained by fitting of experimental data with theoretical dependences following from equations (1) and (2) is shown in the inset of figure 3. The result shows good agreement with the equation and confirms the FMR nature of the signals S_1 and S_2. The effective magnetization $4\pi M_{\mathrm{eff}}$ estimated using the α dependence of the acoustic mode is 3.82 kOe, close to that for $x = 0.4$ single crystal [18].

Figure 4 shows the temperature dependence of the resonance fields for two resonance modes. Both the acoustic mode and the antiparallel optical mode shift to lower field as temperature decreases. The separation in resonance field increases with increasing temperature and then decreases rapidly with the vanishing of the 2D FM correlation above 210 K, which can be seen more clearly from the inset of this figure. The decrease of the field separation below 220 K indicates a weakening of the AFM inter-layer coupling with decreasing temperature. Below T_C, the optical peak merges into the main acoustic peak and the field separation goes to zero, which indicates disappearance of the AFM coupling below T_C.

The temperature dependence of the intensity ratio between the optical and acoustic modes can be used to qualitatively evaluate the IEC strength. As shown in figure 5, the $I^{\mathrm{optic}}/I^{\mathrm{acoust}}$ ratio initially increases with the appearance of AFM correlations and then decreases as temperature decreases below 220 K, denoting a weakening of the coupling on cooling. The inset shows the intensities of the two resonance modes. The acoustic mode intensity I^{acoust} increases gradually with decreasing temperature and shows a peak with the formation of 2D FM correlation, and then increases gradually with further decreasing temperature and saturates near the Curie temperature T_C. Meanwhile, the optical mode intensity I^{optic} increases on cooling and then decreases monotonically with the formation of 2D FM correlations within MnO_2 bilayers, and goes to zero rapidly close to 120 K. These results are consistent with the neutron scattering studies.

To support the final conclusion exactly, a quantitative analysis of the inter-layer coupling may be needed. Unfortunately, the quantitative data for the inter-layer coupling cannot be simply obtained from the FMR spectra. There have been theoretical approaches developed for the determination of inter-layer coupling J_{inter} in the trilayer system from the FMR spectra. The coupling is accounted for as an effective field with the Landau–Lifshitz equation, and the strength is given by J_{inter} which has the units of $\mu\mathrm{eV/atom}$ [11, 19, 20]. However, this theory requires *in situ* FMR measurements in the process of preparing the films and cannot be directly used for the bilayered manganites, which cannot be considered as simple trilayers but include infinite layers. Although a quantitative study is not possible, the results obtained from the FMR spectra are qualitatively consistent with those obtained from neutron scattering and helpful for understanding the inter-bilayer as well as the intra-bilayer spin and charge dynamics and the CMR effect in bilayered manganites.

4. Conclusion

In summary, the inter-layer coupling in a bilayered manganite $La_{1.24}Sr_{1.76}Mn_2O_7$ ($x = 0.38$) single crystal has been identified from FMR spectra. The observation of the high field optical mode indicates the existence of local AFM inter-layer coupling in the paramagnetic state. The analysis of the temperature dependence of the FMR resonance fields and intensities reveals the evolution of AFM inter-layer coupling with temperature. This observation is qualitatively in agreement with previous neutron scattering studies on a $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.4$) single crystal. Compared to neutron scattering technology, the FMR technology is simple and more convenient in the temperature dependent measurements. Therefore, our study suggests that FMR will provide a useful method for investigation of the inter-layer coupling in bilayered manganites.

Acknowledgments

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