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Effect of epitaxial strain on small-polaron hopping conduction in Pr$_{0.7}$(Ca$_{0.6}$Sr$_{0.4}$)$_{0.3}$MnO$_3$ thin films

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We investigated the epitaxial strain effect on the small-polaron hopping conduction properties in Pr$_{0.7}$(Ca,Sr)$_{0.3}$MnO$_3$ (PCSMO) films. An increase in the carrier localization, as evidenced by the enhancement of the small-polaron activation energy E_A in the high temperature region, was obtained by increasing the epitaxial lattice strain in either the tensile or compressive strained film. Furthermore, it was found that the magnitude of E_A, and thus the carrier localization, strongly depends on the sign of the lattice strain, which explains the diverse percolative transport behaviors in PCSMO films with different types of strains. Meanwhile, similar dependencies on the strain of the films were also obtained for the electron-phonon interaction, characterized by the calculated small-polaron coupling constant. Our results reveal that the type of lattice strain plays a crucial role in determining the degree of localization of charge carriers and the electron-phonon coupling strength, which is important for understanding the lattice strain-induced various novel properties related to phase separation and percolation behaviors. © 2015 AIP Publishing LLC.
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Recently, the coexistence and competition of the charge-ordered insulating and ferromagnetic (FM) metallic phases in mixed valence manganites have attracted much attention due to the strong electron–phonon (e–ph) coupling.[1] It is known that the strong electron-lattice (or electron-phonon) coupling would cause the degeneracy and splitting of the singly occupied e_g level in Mn^{3+} accompanied with a distortion of MnO$_6$ octahedra, which are generally termed as Jahn-Teller (JT) distortions. Such effects lower the energy of occupied state but at a cost in lattice energy. It is generally agreed that the net energy gain due to the JT distortion can bind a charge carrier to its site, which plays a key role in various remarkable properties like metal-insulator transition (MIT), charge ordering (CO), phase separation (PS), etc.[2]

A strong electron–phonon coupling may trap the e$_g$ electron in a given Mn orbital due to the potential minimum produced by the Jahn-Teller lattice distortion, which results in the localization of charge carriers. If an e$_g$ electron is localized at a Mn site in the insulating phase, the symmetry of the surrounding oxygen octahedron is lowered from cubic to tetragonal. Such distorted octahedra stack in particular, patterns, often characterized as insulating charge and orbital ordering[3] and accompanied with the local lattice distortions which is absent in the metallic phase with delocalized e$_g$ electrons. Changing the localization of carriers through local lattice distortion will break the subtle balance between competing insulating phase and metallic phase, leading to large changes in the material properties related to phase separation and percolation behaviors. Experiments have demonstrated that the anisotropic strain-field can induce an preferential orbital occupation[4,5] and promote the

growth of uniaxially elongated FM phases[6–8] due to the variation of localization of carriers through JT interaction. As a result, an anisotropic percolation[4,8] and even an abnormal percolation with negative thermal hysteresis[9] were observed in the Pr$_{0.7}$(Ca,Sr)$_{0.3}$MnO$_3$ film at the metal-insulating transition.

Meanwhile, the strong e-ph coupling can result in the formation of polarons,[10,11] which is strongly related to the association of the localized e$_g$ electrons with the lattice distortion. Actually, in most manganite systems, the carriers, together with its associated lattice distortion, are comparable in size to the cell parameter since the localization is enhanced by the JT distortion of the MnO$_6$ octahedron. Thus, a small or Holstein polaron is energetically favorable[10] and play a key role in the conduction mechanism in the high-temperature regime, where magnetic correlations become negligible and charge-lattice interplay dominates.[11,12] In manganite thin films, strong polaronic effects are expected at temperature region above MIT, $T > T_{MI}$, and reflect the localization of the charge carriers. Hence, the behavior of the resistivity in the high temperature paramagnetic (PM) regime is an important probe of the degree of carrier localization. By investigating the hopping characteristics of the small polaron, one can obtain the information of e$_g$ electron localization and the electron-phonon coupling strength, which is crucial for understanding various novel properties related to the phase separation.[3]

On the other hand, the MnO$_6$ octahedra (the Mn-O-Mn bond angle and bond length) in the film can be manipulated by the in-plane biaxial strain introduced by the lattice mismatch between the film and the single crystal substrate. Thus, the degree of the electron localization and the e$_g$ electron bandwidth will alter due to the modification of the lattice energy from MnO$_6$ distortions. As a result, the physical

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properties of manganite films, such as MIT and its temperature, charge ordering, and phase separation, can be largely modulated by the lattice strain.[2,9,13–16] Experimental researches indicated that the $Pr_{0.7}(Ca,Sr)_{0.3}MnO_3$ films undergoing in-plane tensile and compressive strains show different phase separation features.[17] Although both the in-plane tensile- and compressive-strained films show a similar shift of MIT temperature towards low temperature compared to the bulk due to the reduced bandwidth, the tensile-strained films exhibit much wider thermal hysteresis gap and lower MIT temperature than the compressive-strained ones under the same level of strains, indicating different percolation characteristics.[17] It is assumed that such extremely diverse behaviors may be related to the specific localization of e_g electrons resulting from different Jahn-Teller distortion and electron-phonon coupling in the strained films. However, no distinct characterization of the relation between the different type of strains and the localization of charge carriers as well as the electron-phonon coupling has been reported so far. Considering that the formation of small polaron at high temperature is strongly connected to the localization of charge carriers and electron-phonon coupling, it would be very interesting to investigate the behaviors of high temperature polaronic effect under various epitaxial strain, which would provide a good way to reveal the intrinsic relationship between the sign/type dependency of the strain effect and the carrier localization as well as electron-phonon coupling within the framework of polaron hopping.

In this letter, we investigate the influence of the type of the epitaxial lattice strain (magnitude and sign) on high-temperature conduction properties and thus the conduction mechanism of epitaxial $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ (PCSMO) films by measuring the transport properties of the films on various substrates. Four kinds of (001)-orientation single crystals with different lattice constant, $LaAlO_3$ (LAO, 3.788 Å), $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ (LSAT, 3.868 Å), $NdGaO_3$ (NGO, 3.853 Å), and $Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O_3$ (PMN-PT, 4.017 Å), are chosen as substrates to introduce various lattice strains in the films. The data show that the high-temperature resistivity of the films can be well fitted by the adiabatic small-polaron model. We find that the small-polaronic activation energy E_A increases with the increase of both the in-plane tensile and compressive strain, which indicates the enhancement of the charge carrier localization. Meanwhile, strong e-ph coupling in present case was demonstrated by the large polaron coupling constant (γ_p) and the giant ratio of polaron mass (m_p) to rigid lattice effective mass of electrons (m^*) estimated from the fitting of the temperature dependent resistivity. Similar strain dependences were further found for γ_p and m_p/m^*, which indicates that the intensified e-ph interaction should be also responsible for the observed increase of E_A. More interestingly, we found that the magnitudes of the obtained activation energies in the case with tensile strains are larger than the ones in the case with same level of compressive strains. These results reveal that the type of lattice strain plays a crucial role in the localization of carriers and thus the polaronic hopping. Furthermore, we find that the value of γ_p is larger for the film under tensile strain compared with the film under compressive one, which further indicates that the sign of lattice strain play a key role in affecting the electron-phonon interaction.

The PCSMO films were fabricated by pulsed laser deposition (PLD), a KrF excimer laser (248 nm) with a pulsed energy of 300 mJ, and a repetition of 1 Hz was used. The deposition was performed at a substrate temperature of 700 °C and an oxygen pressure of 67 Pa. After the deposition, the samples were cooled down to room temperature in a pure oxygen atmosphere of 1 atm. The thickness of the films was controlled to be 30 and 100 nm, respectively. The crystalline structure and out-of-plane lattice parameters were determined by means of X-ray diffraction (XRD) using Cu–Kα radiation. The transport properties were measured by using the superconducting quantum interference device (SQUID)–VSM, equipped with a home-made electric-measurement module for performing four-point electrical resistance measurements. The XRD θ-2θ scans for the (001)-PCSMO films demonstrate that all films are highly oriented along [001] direction and no other impurity phases or textures are observed (Fig. 1). It was determined from the diffraction patterns that the films on LAO and NGO undergo out-of-plane tensile strain. Typically, the values of 1.63% and 0.37% are extracted for the 30 nm films on LAO and NGO, respectively. The corresponding in-plane strains are calculated as −1.17% and −0.27%, respectively, by Poisson relation $\varepsilon_{zz} = -2\nu/(1-\nu)\varepsilon_{xx}$ using $\nu = 0.41$.[18] Whereas the films on LSAT and PMN-PT exhibit out-of-plane compressive strains (−0.82% for the 30 nm film on LSAT and −1.01% for the one on PMN-PT) and in-plane tensile strains (0.59% for the former and 0.73% for the latter). Detailed results for all films are summarized in Table I.

The temperature dependence of resistivity was measured during warming process over a temperature range between 20 and 300 K with a step of 2.5 K after the sample was cooled down from 300 K to 20 K. It is generally accepted that the polaron hopping behavior in hole-doped manganites at temperatures above T_{MI} is due to the strong e-ph interaction.[19] For present PCSMO films, we found that the resistivity data are dominated by the thermally activated hopping of small polarons in the high temperature ($T > T_{MI}$) insulating phases and can be well described by the Emin-Holstein adiabatic small polaron hopping (SPH) model.[20] According to the Emin–Holstein model of SPH, the resistivity in the adiabatic limit[21–24] is given by $\rho(T) = AT\exp(E_A/k_BT)$, where K_B is the Boltzmann constant, and $A = 2k_B/(3ne^2a^2\nu_{ph})$ is a constant that depends on the polaronic concentration n, the hopping distance a, and the frequency of the longitudinal optical phonon ν_{ph}. E_A is the activation energy for the polaron hopping, and approximately half of the polaron binding energy in the adiabatic limit.

Figure 2 shows the inverse temperature dependence of $ln(\rho/T)$, where a clear linear dependence at temperature above $\theta_D/2$, with θ_D being the Debye temperature, is observed for all studied films. The solid lines in the plot are linear fits to the experimental data. The value of Debye temperature can be extracted from the temperature where the linearity of the $ln(\rho/T)$ Vs. $1/T$ curve (Fig. 2) disappears in the high temperature region. The Debye temperatures θ_D were estimated as ~320.4 K and ~320.6 K (~335.4 K and 325.4 K) for the 30 nm (100 nm) films on LAO and NGO, respectively, which undergo in-plane compressive strain. While for the 30 nm (100 nm) films on LSAT and PMN-PT
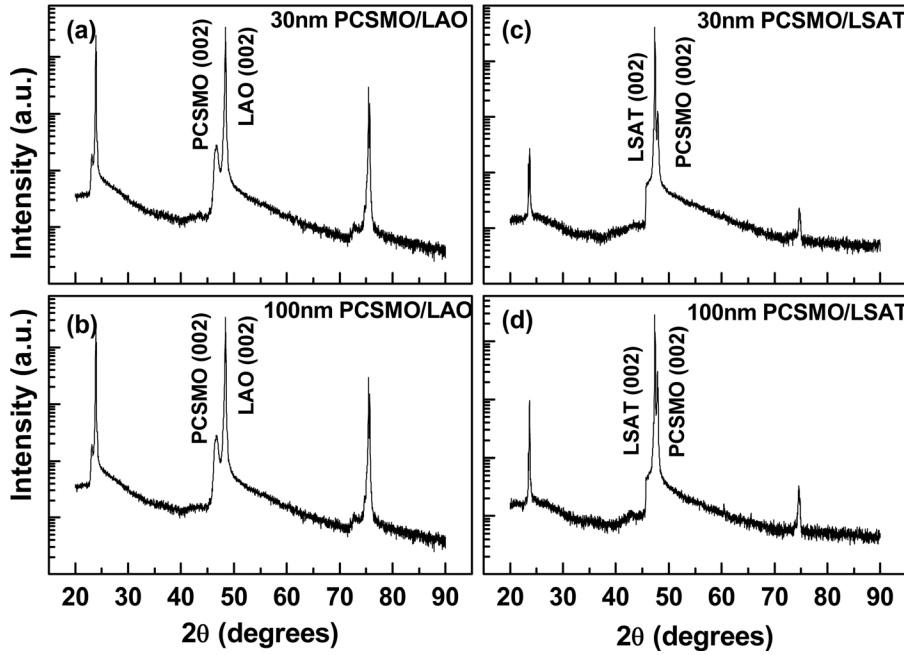
FIG. 1. Typical X-ray diffraction patterns of $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ films on LAO and LSAT substrates. (a) 30 nm and (b) 100 nm films on LAO, and (c) 30 nm and (d) 100 nm films on LSAT.

TABLE I. Lattice strains, MIT temperature T_{MI}, Debye temperature θ_D, and optical phonon frequency ω_0 for $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ films.

Substrate	Film thickness (nm)	Out-of-plane strain (%)	In-plane strain (%)	T_{MI} (K)	θ_D (K)	ω_0 (Hz)
LAO	30	1.63	−1.17	117.3	320.4	6.7×10^{12}
	100	1.58	−1.14	114.8	335.4	7.0×10^{12}
NGO	30	0.37	−0.27	142.3	320.6	6.7×10^{12}
	100	0.33	−0.24	142.3	325.4	6.8×10^{12}
LSAT	30	−0.82	0.59	129.8	305.4	6.4×10^{12}
	100	−0.70	0.50	132.3	315.4	6.6×10^{12}
PMN-PT	30	−1.01	0.73	89.8	280.4	5.8×10^{12}
	100	−0.82	0.58	119.4	325.8	6.8×10^{12}

with in-plane tensile strain, the corresponding values of ∼305.4 K and ∼280.4 K (∼315.4 K and 325.8 K) were obtained. Furthermore, the optical phonon frequency can be estimated from the relation $h\omega_0 = k_B\theta_D$. All obtained results of ω_0 and θ_D are also listed in Table I. These values agree with previously reported Debye temperature in the epitaxial films of $Pr_{0.65}(Ca,Sr)_{0.35}MnO_3$ and $Pr_{0.5}Ca_{0.5}MnO_3$ (Refs. [25] and [26]) but are much smaller than those obtained in the polycrystalline samples.[19]

The E_A was calculated from the fitting parameter and its dependence on the strain was depicted in Fig. 3(a). It can be found that E_A increases with both the magnitudes of in-plane compressive and tensile strain increase, implying that the

lattice strain has a significant effect on the e_g electron localization and thus the polaron hopping in these films. It is known that the lattice strain could change the bending condition of the Mn-O-Mn bond and induce compression or elongation of MnO_6 octahedra. Thus, the JT distortions are enhanced and the electrons tend to become more localization, resulting in the enhancement of the polaronic hopping potential barrier and, hence, the increase in the activation energy. Such enhanced localization effect also leads to a reduction in T_{MI} for films with higher in-plane strain, as shown in Table I. Meanwhile, one can find that the films of 100 nm have higher E_A than the ones of 30 nm. Such enhancement of activation energy in the thicker film was
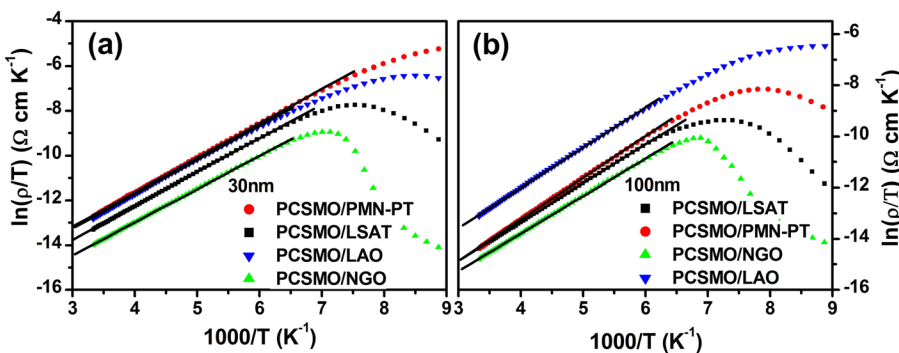

FIG. 2. Variation of $ln(\rho/T)$ of $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ films with thickness of (a) 30 nm and (b) 100 nm on various substrates as a function of inverse temperature. Solid lines are the best fit to the SPH model, viz., $\rho(T)/T = A\exp(E_A/k_BT)$.
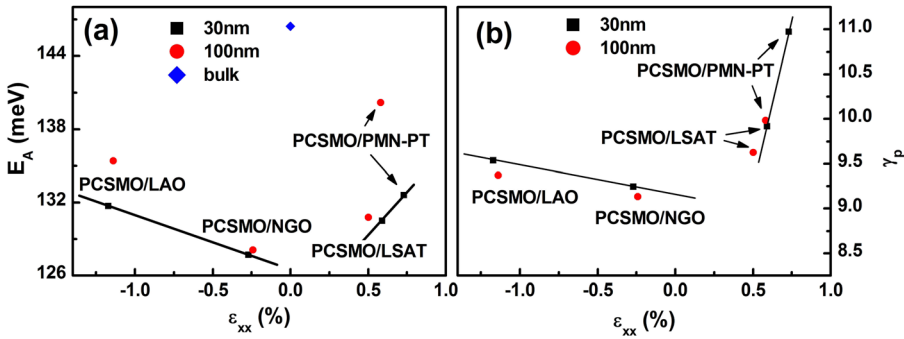
FIG. 3. Dependence of (a) the E_A and (b) the electron-phonon coupling constant on the in-plane lattice strain in $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ films with thickness of 30 and 100 nm. Lines are guides to the eyes.

also observed in low doped $(La,Sr)MnO_3$ (Ref. 27) and $(La,Pr)Ca_{0.3}MnO_3$ films.[28] This anomalous phenomenon should be ascribed to the partial relaxation of the lattice strain due to increased thickness. It is known that the strain relaxation can generate a variety of lattice defects such as dislocations, oxygen vacancies, stacking faults, and so on. These lattice defects enhance the JT distortion and thus strengthen the localization of charge carries, resulting in the enhancement of the activation energy. In present 100 nm film, the impact from lattice defects may be larger than the one from strain reduction. Thus a larger activation energy in the partially relaxed film can be expected. Moreover, the activation energy for the bulk polycrystalline was also calculated and plotted in Fig. 3(a). The value of $E_A \sim 145.4$ meV is larger than the one of strained films but comparable to a polycrystalline with similar composition $Pr_{0.65}Ca_{0.2}Sr_{0.15}MnO_3$ ($E_A \sim 148.02$ meV).[19] The crystalline quality difference between the epitaxial film and polycrystalline bulk, as well as the different defective microstructure should be responsible for the larger activation energy in bulk.[20] In fact, similar phenomenon was also observed in $Pr_{1-x}Ca_xMnO_3$ films and bulk,[26] where the bulk material and annealed films have much larger activation energy than the as-prepared films due to the possible differences of defective microstructures.

More interestingly, the magnitudes of the obtained activation energies are different for the films with compressive and tensile strain. One can distinguish that, for the same level of lattice strains, the tensile strain leads to a bigger E_A in the strained film than the compressive one. This result implies that the polaronic hopping depends on the sign of lattice strain. According to the Emin–Holstein model of SPH, the activation energy $E_A = \varepsilon_0 + E_H - J$, where ε_0 is the energy difference between identical lattice distortions with and without the hole, J is the transfer integral, and E_H is one-half of the polaron binding energy.[3,21,29] Previous works revealed that $\varepsilon_0 \approx$ few meV $\ll E_A \approx 100–200$ meV for the manganites,[3,29] in accordance with the assumption of small polarons where the local lattice distortion is comparable in size to the cell parameter. For the present PCSMO film, the lattice strain would enhance the Jahn-Teller distortion and thus the localization of carriers, resulting in an increase of the polaron binding energy E_H. Meanwhile the transfer integral J is a very sensitive function of the inter-site separation which could be represented by the Mn-O-Mn bond length and angle in the small polaron hopping model. The in-plane tensile strain tends to stretching the MnO_6 octahedra in the plane and compressing it along the out-of-plane direction, resulting in an increase of bond length and thus an increase

of the small polaron hopping distance. As a result, the increased tensile strains will lead to a decrease in transfer integral J, while the compressive strain does the opposite.[2] However, previous researches have shown that $J \ll E_H$, if the condition $T < E_H/k_B$ (~ 1200 K in the manganites system) is satisfied.[29] Thus, the changes of J due to the increased in-plane lattice strain should be much smaller than the increase of E_H from the enhancement of carrier localization. As a result, the E_A increases with the strength of both tensile and compressive strain except that the compressive case leads to a smaller slope in the strain dependent E_A than the tensile case.

Furthermore, previous investigations on $La_{0.67}Ca_{0.33}MnO_3$ films[27,30] indicated that the tensile strain causes stronger electron localization than the compressive one. In fact, the different types of strain will cause a different MnO_6 distortion. Such different local lattice distortions would produce different level of potential minimum and tend to trap the e_g electron in different orbitals.[5] As a result, the localization of charge carriers and thus E_A behaves different under the conditions of tensile and compressive strain, which should be the main reason of the diverse characteristics of the percolation transport behaviors observed in PCSMO films with different types of strain.

To further account for the origin of the increased activation of small polaron in the strained films, we made an effort to calculate the value of small polaron coupling constant γ_p, which is a measure of the e-ph interaction strength. The values of γ_p are calculated from the relation $\gamma_p = 2E_H/\hbar\omega_0$.[31,32] For a rough estimation, we use $E_H \cong E_A$ in the adiabatic limit,[29] and the calculated values are given in Table II. It is seen that the obtained γ_p for all samples varies from 9.1 to 11.0, suggesting very strong e-ph interaction in present system.[19,32] The ratio of polaron mass (m_p) to rigid lattice effective mass of electrons (m^*) was further calculated using

TABLE II. Parameters of activation energy E_A, e-ph coupling constant γ_p, and the ratio of polaron mass to rigid lattice effective mass of electrons m_p/m^* for $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ films.

Substrate	Film thickness (nm)	E_A (meV)	γ_p	m_p/m^*
LAO	30	131.7	9.5	13359.7
	100	135.4	9.4	12088.4
NGO	30	127.7	9.2	9897.1
	100	128.1	9.1	8955.3
LSAT	30	130.5	9.9	19930.4
	100	130.8	9.6	14764.8
PMN-PT	30	132.6	11.0	59874.1
	100	140.2	10.0	22026.5

$m_p = m^* \exp(\gamma_p)$[31] and the results are also shown in Table II. The large values of m_p/m^* further confirm the strong e-ph interaction.[19] Furthermore, one can find that γ_p as well as m_p/m^* increases with the increase of either the tensile or the compressive in-plane strain (see Fig. 3(b)), which indicates an enhancement of e-ph interaction in the strained films. Such intensified e-ph interaction will strengthen the charge localization and leads to an increase in the energy required to liberate a free carrier and thereby the activation energy of polaron increases. Furthermore, one can find that the value of γ_p is larger for the film under tensile strain compared with the film under compressive one, which further indicates that the sign of lattice strain plays a key role in affecting the electron-phonon interaction.

In summary, the strain dependent high temperature transport properties of $Pr_{0.7}(Ca_{0.6}Sr_{0.4})_{0.3}MnO_3$ films were illustrated by considering the adiabatic small polaron hopping conduction above MIT temperature. The small-polaronic E_A, extracted from the fitting of temperature dependent resistivity in the high-temperature region, was found to increase with increasing the in-plane strain. The effect of lattice strain on JT distortion and hence the localization of charge carries was used to explain such behavior. Furthermore, a strain sign dependency of E_A was distinguished by comparing the results of both tensile and compressive strained-films. Meanwhile, large e-ph coupling constants (γ_p) and giant ratio of polaron mass (m_p) to rigid lattice effective mass of electrons (m^*) were further calculated using the small polaron hopping model, demonstrating a similar strain dependence. These results suggest that the type of lattice strain (magnitude and sign) plays a crucial role in determining the degree of localization of charge carriers and the strength of e-ph interaction, which should be closely related to the different characteristics of phase separation and percolation behaviors under different strains.

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