# Nonequilibrium superconductivity under spin-polarized quasiparticle currents in perovskite ferromagnet-insulator-superconductor heterostructures

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(Received 8 July 1999)

We report phenomena manifesting nonequilibrium superconductivity induced by spin-polarized quasiparticles in perovskite ferromagnet-insulator-superconductor (F-I-S) heterostructures. Measurements of the critical current $J_c$, using a pulsed current technique to minimize Joule heating, reveal a monotonic *increase* with increasing insulator thickness; and for F-I-S with *thin* insulating barriers, a slight *increase* in $J_c$ is observed under small injection currents $I_m$ from the ferromagnet, followed by a strong *suppression* of $J_c$ under large $I_m$. In contrast, no effect of injection on $J_c$ can be detected in the N-I-S control sample (N: nonmagnetic metal). The spin diffusion time and length in cuprate superconductors are also estimated and compared with our experimental results. [S0163-1829(99)05638-6]

## I. INTRODUCTION

Many phenomena associated with nonequilibrium superconductivity have been investigated extensively since the 1970s.[1–8] These studies have primarily focused on the effects of simple quasiparticle injection and extraction,[1–4] or spin-polarized quasiparticle transport,[5,6] in conventional *s*-wave superconductors. In the case of simple quasiparticles, both enhancement and suppression of superconductivity have been observed, and the phenomena can be generally described in terms of a nonequilibrium energy $(T^*)$ and a net quasiparticle charge density $(Q^*)$.[1–4] For the injection of spin-polarized quasiparticles, experimental studies have demonstrated that electrical currents from ferromagnets may become spin polarized,[6–8] and the spin-polarized current may tunnel through an insulating barrier into a conventional superconductor,[6,8] resulting in suppression of superconductivity due to the breaking of time-reversal symmetry of the Cooper pairs, which is analogous to the pair-breaking effect of localized magnetic moments.[9–11] Recently, the concept of spin injection has been extended to perovskite ferromagnet-insulator-superconductor (F-I-S) heterostructures,[12,13] and suppression of the superconducting critical current has been reported in cuprate superconductors by injecting dc currents from the underlying ferromagnetic manganite film. This suppression has been attributed to magnetic pair breaking. However, questions remain in that significant Joule heating in those experiments ($\sim$ 10 mW/cm$^2$ in Ref. 12 and up to $\sim$ 1 W/cm$^2$ in Ref. 13), which are generated by passing a large dc current through the resistive ferromagnetic underlayer, may result in dominating thermally induced pair breaking effects. Furthermore, none of those earlier reports has provided either microscopic evidence for pair breaking or estimates for important physical quantities such as the spin diffusion time or length.

In this work, we report phenomena in perovskite ferromagnet-insulator-superconductor (F-I-S) heterostructures that are manifestations of nonequilibrium superconductivity induced by spin-polarized quasiparticles. A pulsed current technique and *in situ* thermometry are employed to ensure that Joule heating is limited to less than 10 mK, and therefore can be neglected. F-I-S heterostructures with two different compositions and various insulating barrier thicknesses have been investigated. In addition, control samples of N-I-S heterostructures (N: nonmagnetic perovskite metal) and plain superconducting films are also studied for comparison. In the absence of any significant Joule heating, several phenomena that had not been observed by those measurements using large dc currents[12,13] are revealed, and are attributed to the dynamic pair-breaking effects of spin-polarized quasiparticle currents.

## II. EXPERIMENTAL

The *c*-axis-oriented heterostructures used in this work include the following:

$La_{0.7}Ca_{0.3}MnO_3$ (100 nm)/YSZ (1.3 nm)/
$YBa_2Cu_3O_7$ (100 nm) (LCMO/YSZ/YBCO),

$La_{0.7}Sr_{0.3}MnO_3$ (100 nm)/$SrTiO_3$ (2 nm)/
$YBa_2Cu_3O_7$ (100 nm) (LSMO/STO/YBCO),

$La_{0.7}Sr_{0.3}MnO_3$ (100 nm)/$SrTiO_3$ (10 nm)/
$YBa_2Cu_3O_7$ (100 nm) (LSMO/STO′/YBCO),

$LaNiO_3$(100 nm)/$SrTiO_3$ (2 nm)/
$YBa_2Cu_3O_7$ (100 nm) (LNO/STO/YBCO),

 

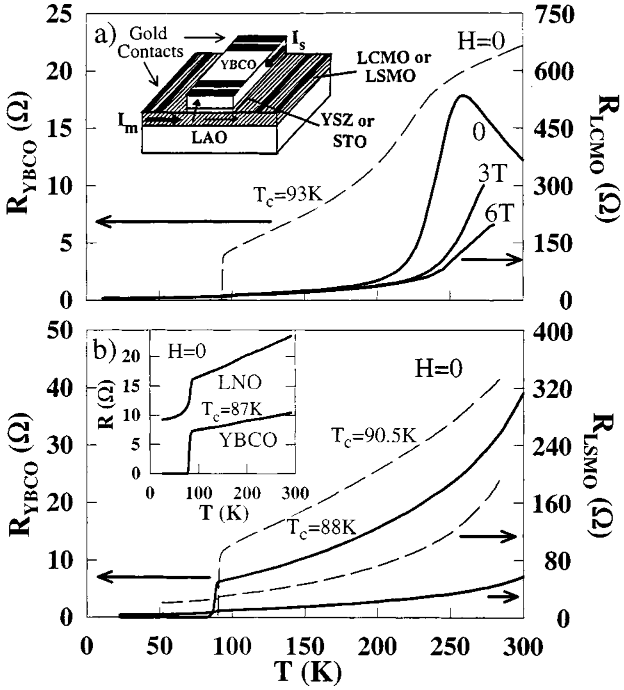


FIG. 1. (a) Resistance ($R$) vs temperature ($T$) curves of the YBCO (left axis) and LCMO (right axis) in the LCMO/YSZ/YBCO heterostructure. The inset illustrates schematics of such heterostructures and the electrical contact configurations. (b) The $R$ vs $T$ curves of the YBCO (left axis) and LSMO (right axis). The solid lines denote the data for LSMO/STO/YBCO, and the dashed lines are the data for LSMO/STO′/YBCO.

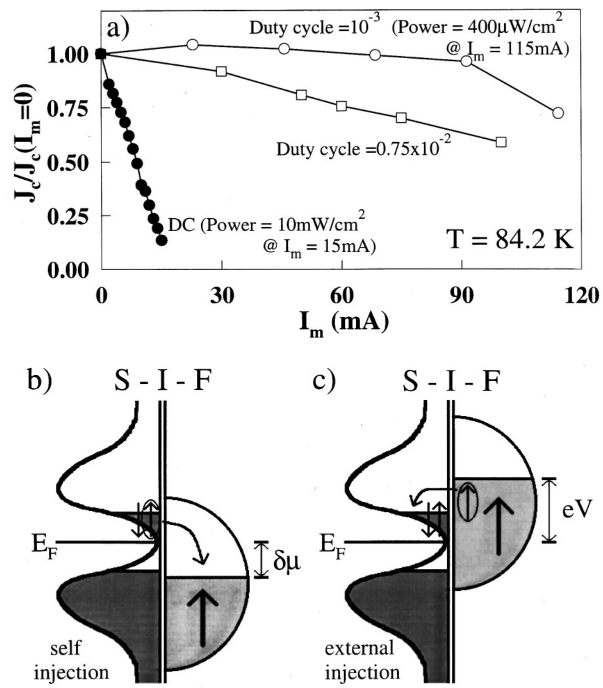


FIG. 2. (a) Comparison of the normalized critical current density $J_c/J_c(I_m=0)$ vs $I_m$ data of LCMO/YSZ/YBCO for dc and pulsed injection current ($I_m$) at a nominal temperature $T=84.2$ K. (b) Schematic illustration of the ''self-injection'' of spin-polarized quasiparticles near the F/S interface. (c) Schematics of external injection of spin-polarized quasiparticles from F to S. Here $E_F$ denotes the Fermi energy of the superconductor and $V$ is the external bias voltage across the F/S interface.

where YSZ denotes yttrium-stabilized zirconia. These samples are fabricated using pulsed laser deposition on $LaAlO_3$ (100) (LAO) substrates which are 6 mm×6 mm in size. The procedure involves first deposition of the ferromagnetic manganite or nonmagnetic nickelate layer on an LAO substrate, then a thin insulating barrier and YBCO on top of the masked manganite to yield a strip 6 mm×2 mm. The heterostructures are slowly cooled to room temperature in 500 Torr oxygen. At least two samples of each heterostructure are grown together for different characterizations and measurements. X-ray photoelectron spectroscopy (XPS) has been performed on the bilayers to ensure that no visible reaction occurs during the growth process.

For the electrical transport measurements, 8 gold contacts, 4 on YBCO and 4 on the magnetic layer, are sputtered onto each heterostructure, as illustrated in the inset of Fig. 1(a). The resistance ($R$) as a function of the temperature ($T$) for the constituent layers of each sample is monitored with a small direct current (3 $\mu$A), to ensure negligible Joule heating. The results on LCMO/YSZ/YBCO and LSMO/STO(STO′)/YBCO are shown in Figs. 1(a) and 1(b), respectively, and those on LNO/STO/YBCO are shown in the inset of Fig. 1(b). Due to a finite junction resistance ($R_J$) across the F-I-S and N-I-S heterostructures,[14] the data are displayed in resistance rather than resistivity. However, we note that for all heterostructures, the normal state resistivity $\rho$ of the superconducting layer is $\rho \approx 60\,\mu\Omega$ cm at the superconducting transition temperature $T_c$, comparable to the in-plane resistivity of high-quality YBCO single crystals.[15] In addition, scanning tunneling spectroscopy (STS) studies on the YBCO layer of these heterostructures[16] have revealed spectra comparable to those of YBCO single crystals[17] and consistent with the $d$-wave symmetry.[17,18] Resistance measurements are also performed on the ferromagnetic and nonmagnetic layers, as shown in Fig. 1(a) for the LCMO layer at $H=0,3,6$ T, in Fig. 1(b) for the LSMO layers at $H=0$, and in the inset of Fig. 1(b) for LNO layer at $H=0$. The Curie temperatures as well as the resistivities of both the LCMO and LSMO layers of the F-I-S samples are comparable to those of the bulk material and epitaxial films on LAO substrates.[19] Hence, the quality of the constituent layers apparently remains very high in all heterostructures.

The critical current ($I_c$) measurements of the YBCO are performed by means of a pulsed current technique, in order to prevent undesired Joule heating on YBCO from power dissipation in the resistive electrical contacts and the metallic underlayer. To investigate the effect of injection currents ($I_m$), two pulsed current generators which supply currents to YBCO and the metallic underlayer are synchronized and referenced to the same ground. The pulse width is chosen to yield a negligible temperature rise (<10 mK) in the YBCO under the maximum injection currents (~300 mA), as determined by monitoring the changes in the resistance of the ferromagnetic layer during the measurements. The ratio of the pulsed current width ($t_w$) to period ($t_p$) thus chosen is 1:1000, and the measurement of $I_c$ is independent of the pulse width for $10 < t_w < 500\,\mu$s. A typical pulse width used for this work is $t_w = 200\,\mu$s. The critical current $I_c$ is empirically defined as the current at which the voltage across the

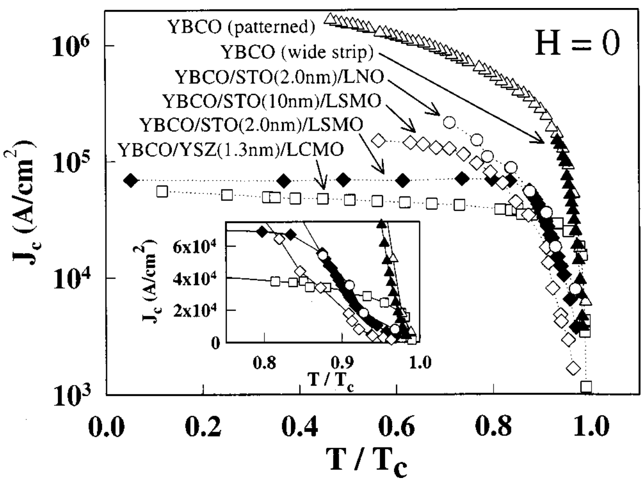


FIG. 3. Comparison of the zero-field critical current density ($J_c$) of YBCO and perovskite heterostructures as a function of the reduced temperature ($T/T_c$). Details of $J_c(T)$ near $(T/T_c)=1$ are shown in the inset.

length of YBCO is 3 $\mu$V, measured with a digital oscilloscope, and $J_c$ is obtained by dividing $I_c$ by the cross-sectional area ($2\ \mathrm{mm}\times 100\ \mathrm{nm}$) of YBCO.

## III. RESULTS

In Fig. 2(a) we compare the $J_c$ vs $I_m$ data of LCMO-YSZ-YBCO taken by both the pulsed current technique and the dc technique. For the same nominal temperature of $T=84.2$ K indicated by a carbon-glass temperature sensor adjacent to the sample, a large suppression of the critical current $I_c$ is observed for a dc current $I_c \leqslant 15$ mA, and the corresponding power dissipation is $\sim 10$ mW/cm$^2$ at $I_m = 15$ mA. In contrast, the suppression of $J_c$ by $I_m$ becomes much less significant under a pulsed current with $t_w = 200\ \mu$s and $t_p = 200$ ms. This comparison underscores the necessity of employing a pulsed current technique to reveal genuine magnetic pair-breaking effects in F-I-S heterostructures.

The temperature dependence of $J_c$ in YBCO for all heterostructures, measured using the pulsed current technique and for $I_m = 0$, are shown in the main panel of Fig. 3, with details of the data near $(T/T_c)=1$ illustrated in the inset. For comparison, the $J_c$ vs $T$ curves of two plain YBCO/LAO films grown under the same condition, one patterned into a ($10\ \mu\mathrm{m}\times 100\ \mu\mathrm{m}$) narrow bridge and the other a ($2\ \mathrm{mm}\times 6\ \mathrm{mm}$) strip, are also included in Fig. 3. Three features are noteworthy. First, the $J_c$ of F-I-S heterostructures at low temperatures *increases* monotonically with increasing barrier thickness. Second, the low-temperature $J_c$ of the N-I-S sample is *higher* than all the F-I-S heterostructures, and those of the two plain YBCO films are comparable and are the highest among all samples. Third, the $J_c$ values of the thin-barrier F-I-S samples are nearly temperature *independent* at low temperatures, in sharp contrast to the strong temperature dependence of $J_c$ in plain YBCO films.

In the presence of a finite $I_m$, the corresponding critical current $I_c$ through YBCO is obtained from the relation $I_c = (I_c^+ - I_c^-)/2$, where $I_c^+$ and $I_c^-$ are defined as the currents at which the voltages along the YBCO film are $+3$ and $-3\ \mu$V, respectively. We note increasing asymmetry between $I_c^+$ and $I_c^-$ with increasing $I_m$, consistent with the phenomena of nonequilibrium superconductivity.[1–3,15] Furthermore, for $I_m$ values that incur little suppression of $J_c$, we find that $I_m \approx (I_c^+ - I_c)/2 \approx (I_c - I_c^-)/2$, which is consistent with a nearly perfect transmission of $I_m$ from F to S. Interestingly, $J_c$ of the *thin-barrier* F-I-S samples exhibits a slight *increase* with $I_m$, followed by a pronounced *suppression* of $J_c$ at sufficiently large $I_m$ near $T_c$, as shown in the main panel of Fig. 4(a) for LSMO/STO (2 nm)/YBCO, and in the inset for the LCMO/YSZ (1.3 nm)/YBCO. In contrast, we find no visible suppression of $J_c$ by $I_m$ for LSMO/STO (10 nm)/YBCO and for the control sample LNO/STO (2 nm)/YBCO at all temperatures, as shown in the inset and the main panel of Fig. 4(b), respectively.

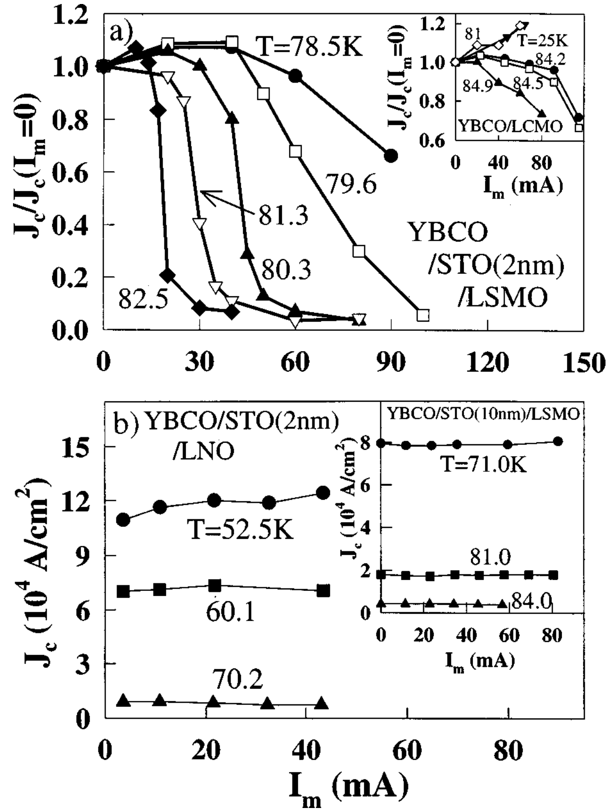


FIG. 4. (a) The $(J_c/J_c^0)$ vs $I_m$ isotherms of LSMO/STO/YBCO. Here $J_c^0(T)$ denotes the critical current density at $T$ and for $I_m = 0$. The inset shows the corresponding results for LCMO/YSZ/YBCO. (b) The $J_c$ vs $I_m$ isotherms of LNO/STO/YBCO and LSMO/STO′/YBCO are shown in the main panel and the inset, respectively.

## IV. DISCUSSION

In the context of no degradation in the quality of the constituent layers, we first discuss the implication of strongly suppressed low-temperature $J_c$ in F-I-S samples with $I_m = 0$. (See Fig. 3.) We note that the predominant $d_{x^2-y^2}$ pairing symmetry of YBCO (Refs. 17,18) allows substantial low-energy excitations, and a significant population of quasiparticles already exists in YBCO even at very low temperatures. It is also known that impedance mismatch exists at the interface of dissimilar materials for thermal[20] or electrical transport,[3] so that a chemical potential difference $\delta\mu$ develops if a current flows across the interface. Hence, if an electrical current is applied to S, and if S and F share a common ground, some thermally induced quasiparticles may diffuse from S to F, yielding $\delta\mu > 0$, which assists further quasiparticles diffusion, as illustrated in Fig. 2(b). Noting that the ferromagnetic manganites are half metals with nearly perfect spin polarization,[19,21,22] only quasiparticles with spins parallel to those of the majority carriers in F may diffuse across the interface, leaving behind spin-polarized quasiparticles in YBCO and giving rise to further pair breaking. Therefore, significant suppression of $J_c$ occurs for thin-barrier F-I-S samples at low temperatures even if $I_m = 0$. This ''self-injection'' scenario in F-I-S [see Fig. 2(b)] is further corroborated by the observation of monotonic *increase* of low-temperature $J_c$ with increasing insulator thickness, because increasing interface impedance reduces the transport probability of quasiparticles from S to F,[23] thereby reducing the spin-polarized quasiparticles due to self-injection. In contrast, $J_c$ of the N-I-S sample is the largest among all heterostructures, and is only smaller than that of plain YBCO (see Fig. 3), suggesting a much smaller pair-breaking effect of simple quasiparticles.

While vortex nucleation near the edge of wide YBCO films may also incur suppression of $J_c$, it *cannot* account for the systematic increase in $J_c$ with increasing insulating barriers in F-I-S, because the dimension of YBCO is the same for all heterostructures and for one of the plain films. Furthermore, for a given reduced temperature, $J_c$ is comparable for both the patterned YBCO narrow bridge and the (2 mm×6 mm) wide strip, suggesting that the large suppression of $J_c$ in F-I-S is not attributable to vortex nucleation near the edge of wide YBCO strips in the heterostructures.

The self-injection conjecture also appears consistent with the $J_c$ vs $I_m$ data in Fig. 4(a). That is, quasiparticles due to self-injection would possess a net spin polarization *opposite to* that of the majority carriers in the ferromagnet. Hence, for externally injected spin-polarized quasiparticles with energies larger than the averaged superconducting energy gap,[24] initial *increase* in $J_c$ may occur with $I_m$ due to the *neutralization* of externally injected and self-injected spin-polarized quasiparticles. However, $J_c$ must decrease eventually with further increase in $I_m$ because of the pair-breaking effect of spin-polarized quasiparticles. Such a conjecture is consistent with the experimental data in Fig. 4(a), and is schematically illustrated in Fig. 2(c). In contrast, $J_c$ appears *independent* of $I_m$ in thick-barrier F-I-S samples for the range of currents studied, which may be due to the large interface impedance that reduces the transmission of spin-polarized quasiparticles.[23] In addition, no suppression of $J_c$ with injection can be observed in the thin-barrier N-I-S sample [see Fig. 4(b)], suggesting that *the $J_c$ suppression in F-I-S samples is predominantly due to the pair-breaking effect of spin-polarized quasiparticles*. This conclusion is further supported by our recent scanning tunneling spectroscopy studies,[16] showing substantial increase in the quasiparticle density of states (DOS), which is indicative of pair breaking, only in the thin-barrier F-I-S sample for injection energies $E \sim e(I_m R_J)$ exceeding the averaged $c$-axis superconducting energy gap $\langle\Delta\rangle$. In contrast, no change in the quasiparticle DOS has been observed in the N-I-S sample up to very high $I_m$.[16] The absence of either macroscopic $J_c$ suppression or microscopic quasiparticle DOS increase,[16] due to the injection of simple quasiparticles in N-I-S, suggests that the lifetime and diffusion length of simple quasiparticles are much shorter than those of spin-polarized quasiparticles, because the latter can be relaxed only via spin-orbit or magnetic impurity scattering.

Finally, we suggest that the strong temperature dependence of $J_c$ near $T_c$ is consistent with a diverging spin-polarized quasiparticle diffusion time $\tau_s$.[1–3] This diffusion time may be estimated by the relation $\tau_s \sim 3.7\tau_{ex}k_B T_c/\langle\Delta(T)\rangle$, where $\tau_{ex}$ is the energy relaxation time[3] for spin-polarized quasiparticles. Assuming that $\tau_{ex}$ in the YBCO layer of the F-I-S is primarily determined by the spin exchange interaction, we find $\tau_{ex} \sim (\hbar/E_{ex})$, where $E_{ex} \sim 30\,\mathrm{K}$ is the on-site spin exchange interaction in YBCO.[25] Using an average energy gap $\langle\Delta(T)\rangle \approx \langle\Delta_0\rangle[1-(T/T_c)]^{1/2}$ for the $c$-axis transport, where $\langle\Delta_0\rangle \sim 20\,\mathrm{meV}$,[16,17] we estimate $\tau_s \sim 10^{-12}\,\mathrm{s}$ for $[1-(T/T_c)] \sim 0.01$, and $\tau_s \sim 10^{-13}\,\mathrm{s}$ for $T \to 0$. For a Fermi velocity $v_F \sim 10^5$ m/s and a mean-free path $l_0 \sim 20\,\mathrm{nm}$, the spin diffusion length becomes $l_s \equiv \sqrt{l_0 v_F \tau_s} \sim 80\,\mathrm{nm}$ at $[1-(T/T_c)] \sim 0.01$, and $l_s \sim 10\,\mathrm{nm}$ for $T \to 0$. These estimates suggest that $l_s$ is only comparable to the thickness of YBCO near $T_c$, which accounts for the observed strong suppression of $J_c$ by $I_m$ in the thin-barrier F-I-S samples.

## V. CONCLUSION

In summary, phenomena manifesting nonequilibrium superconductivity in perovskite F-I-S heterostructures are observed with a pulsed current technique that effectively eliminates Joule heating, and are attributed to the dynamic pair-breaking effect of spin-polarized quasiparticles in cuprate superconductors. The $c$-axis spin diffusion length $l_s$ and diffusion time $\tau_s$ are also estimated. Further investigation of the quasiparticle density of states under directional spin injection is in progress to elucidate quantitative properties of nonequilibrium superconductivity and magnetic pair-breaking mechanism in $d$-wave cuprate superconductors.

## ACKNOWLEDGMENTS

This work is jointly supported by NSF Grant No. DMR-9705171, and the Jet Propulsion Laboratory (JPL) Director's Research and Development Fund (DRDF). Part of the research was performed by the Center for Space Microelectronics Technology, JPL, California Institute of Technology, through an agreement with the National Aeronautics and Space Administration. We thank Dr. J. B. Barner at JPL for providing the critical current data on a patterned YBCO epitaxial film, and acknowledge useful discussions with Dr. J. Y. T. Wei (Caltech), Professor Q. Si (Rice University), and Dr. A. W. Kleinsasser (JPL).

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