

Magnon-broadening of exchange-split surface states on lanthanide metals

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Low-temperature scanning tunneling spectroscopy is used to study the electronic structure and the dynamics of d -like surface states on trivalent lanthanide metals from La to Lu. We find that the dynamics of these states, as revealed by their lifetime widths, is dominated by electron-phonon scattering in the occupied region and by electron-magnon scattering in the unoccupied region. We also show that the magnetic exchange splitting of these states scales with the $4f$ spin multiplied by an effective exchange-coupling constant that increases with $4f$ occupancy in an approximately linear way.

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Electronic structure and the dynamics of surface states have recently attracted a substantial amount of attention. For the noble metals, a profound understanding of binding energies and lifetimes of surface states has been achieved, mainly due to improvements in the theoretical and experimental techniques, the latter involving photoemission (PE), inverse photoemission (IPE), time-resolved PE (TRPE), and two-photon PE (2PPE), as well as scanning tunneling spectroscopy (STS).¹ While PE, IPE, and STS measure spectral lifetime widths of electronic states, TRPE and 2PPE are time-resolved techniques that are capable of directly determining relaxation times. On defect-free noble-metal surfaces, the lifetimes of surface states are determined predominantly by electron-phonon (e -ph) and electron-electron (e - e) scattering rates, with the latter including both intraband and interband excitations.²

On the other hand, much less is known on surface-state dynamics in transition metals, particularly in the lanthanides. Across the series, the lanthanide metals have similar conduction-band structures but different $4f$ occupancies, n_{4f} , and hence different $4f$ -spin moments that interact with the ($5d6s$)-conduction electrons and the surface state by exchange interaction. Below specific temperatures, all lanthanide metals with nonvanishing $4f$ moment exhibit long-range magnetic order that causes an exchange splitting of the electronic states.³⁻⁶ For these magnetic transition metals, collective excitations of spin waves (magnons) provide additional decay channels for excited electronic states.⁷ Such electron-magnon (e - m) scattering processes, where an electron undergoes a spin-flip and a magnon is either created or annihilated, however, are difficult to distinguish from e -ph scattering processes since both are expected to exhibit similar temperature dependences.⁸

Recently, the influences of e - m scattering on the electronic lifetimes of various surface states on Fe have been observed by PE and 2PPE experiments.^{9,10} Moreover, the role of e - m coupling in the ultrafast demagnetization process upon optical excitation of the Gd(0001) surface state was revealed in a combined TRPE and nonlinear magneto-optical experiment.¹¹ These observations demonstrate that e - m coupling may play an essential role in ultrafast magnetic recording as well as in high-temperature superconductivity, and they encourage further studies on magnetic surfaces.

We report on a systematic low-temperature STS study of the $5d_{z^2}$ -like surface states on the (0001) faces of trivalent lanthanide metals that exhibit magnetic exchange splittings

into states below and above the Fermi level in the magnetically ordered phases. We find that the dynamics of these states is quite different in the occupied and unoccupied regions, with much larger linewidths (i.e., shorter lifetimes) of the unoccupied minority states as compared to the occupied majority states. These observations reveal that the lifetimes of occupied surface states are mainly determined by e -ph scattering, while those of unoccupied states are dominated by e - m scattering. Furthermore, the magnetic exchange splitting of these states is found to deviate systematically from a sole dependence on the $4f$ spin reported so far,^{5,12} an observation that can be explained by lanthanide contraction.

The experiments were performed in ultrahigh vacuum (UHV) with a low-temperature scanning tunneling microscope (STM) operated at 10 K.⁵ The samples were prepared *in situ* by electron-beam evaporation of the lanthanide metals La($4f^0$), Nd($4f^3$), Gd($4f^7$), Tb($4f^8$), Dy($4f^9$), Ho($4f^{10}$), Er($4f^{11}$), Tm($4f^{12}$), or Lu($4f^{14}$), and deposition on a clean W(110) single-crystal surface kept at room temperature. Subsequent annealing at temperatures between 500 and 1000 K resulted in smooth hcp (0001) films (dhcp in the case of La and Nd) with atomically flat terraces, and local-thickness variations of only a few monolayers (ML).⁵ The average thickness of the deposited film (≈ 30 ML) was monitored by a quartz microbalance. STS spectra were recorded with fixed tip position and switched-off feedback control. The differential conductivity, dI/dU , with I being the tunneling current and U the sample bias voltage, was measured as a function of U by modulating U and recording the induced modulation of I via lock-in technique. dI/dU is known to be proportional to the density of states at the sample surface in good approximation. A modulation amplitude of 1 mV (rms) at a frequency of ≈ 360 Hz was used, with the time constant of the lock-in amplifier set to 100 ms, at a sweep rate of 2 mV/s. To correct for binding-energy shifts due to the finite time constant, the spectra were recorded in the two opposite directions from lower to higher and from higher to lower sample bias. Since both the STM tip and the sample were cooled to (10 ± 1) K, the energy resolution was ≈ 3 meV, corresponding to $3.5 k_B T$.

Figure 1 gives an overview of tunneling spectra of trivalent lanthanide metals in the region of the d_{z^2} -like surface states around E_F , which show up as clearly resolved peaks. For the lanthanides with nonvanishing $4f$ -spin moments, majority and minority exchange-split spin states appear below

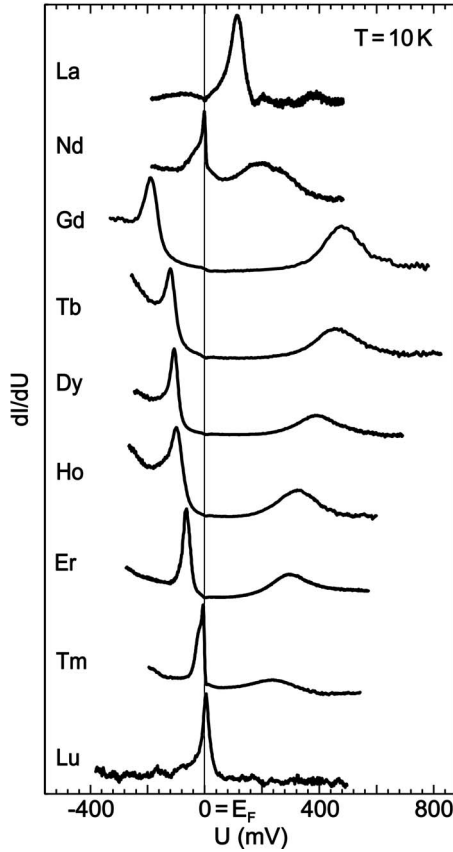


FIG. 1. STS spectra recorded of surface states on trivalent lanthanide metals at 10 K. Single peaks are observed for the nonmagnetic metals La and Lu, while the surface states are exchange split in the case of the other magnetically ordered metals.

and above E_F , respectively. For La and Lu, with empty ($n_{4f}=0$) and completely filled ($n_{4f}=14$) $4f$ shell, respectively, the spectra exhibit a single peak. The strongly peaked surface-state densities probed by STS are a consequence of the high degree of spatial localization,^{13,14} resulting in small dispersions of the surface-state bands, with effective masses $|m^*/m| \gg 1$.^{5,15} In the case of Nd and Tm, with the majority surface states rather close to E_F , additional narrow features directly at E_F show up in the STS spectra that represent many-body resonances due to interaction of the localized surface state with bulk conduction electrons; hence, these resonances exhibit Fano-like shapes.^{16,17}

Due to the small dispersions of the surface-state bands, the surface-state peaks in the STS spectra can be described by Lorentzian lines, with widths Γ that are related to lifetimes τ by $\Gamma = \hbar / \tau$. Only for narrow peaks do the dispersions lead to small deviations from symmetric line shapes. Since the surface-state bands exhibit downward dispersions with band maxima at $\bar{\Gamma}$ (with the exception of Lu, for which an M-shaped dispersion had been calculated¹³), the trailing edges of the peaks in the STS spectra are generally steeper than the leading edges. In order to account for these asymmetries, the peaks were fitted with adequate superpositions of Lorentzian lines.^{5,13} Fit results for energies and linewidths in the present work refer to surface states at the band maximum. In the case of Nd and Tm, the trailing edges of the

TABLE I. Lifetime widths Γ and energies E (relative to $E_F=0$) of exchange-split (0001)-surface states on trivalent lanthanide metals; \uparrow and \downarrow refer to (occupied) majority and (unoccupied) minority states, respectively. The numbers in parentheses represent error bars in units of the last digit.

		E (meV)	Γ (meV)
La		130(5)	50(9)
	\uparrow	-18(5)	70(9)
Nd	\downarrow	190(9)	120(9)
	\uparrow	-191(9)	33(5)
Gd	\downarrow	470(1)	130(9)
	\uparrow	-111(5)	22(5)
Tb	\downarrow	474(5)	180(9)
	\uparrow	-102(5)	19(5)
Dy	\downarrow	402(5)	170(9)
	\uparrow	-90(9)	19(9)
Ho	\downarrow	350(9)	130(9)
	\uparrow	-50(9)	30(9)
Er	\downarrow	318(5)	150(9)
	\uparrow	-19(5)	29(5)
Tm	\downarrow	240(9)	190(30)
Lu		2(1)	5(1)

surface-state peaks are superimposed on the above-mentioned narrow resonances at E_F , with the consequence that the asymmetries of the peaks cannot be determined unambiguously. Therefore, single Lorentzian lines were fitted to these peaks. The error bars that result from a neglect of a dispersion of surface states are less than 10 meV for both energy and width of a given surface state.⁵

Table I summarizes the fit results for energies, E_{\uparrow} and E_{\downarrow} , and widths, Γ_{\uparrow} and Γ_{\downarrow} , of the occupied (\uparrow) and unoccupied (\downarrow) surface states as obtained from a large number of STS spectra for each lanthanide metal. Except for Nd, all studied lanthanide metals order ferromagnetically within the surface plane, and the sample temperature was far below the respective highest magnetic ordering temperature in each case.¹⁸ Hence, the spin polarization at 10 K should be close to 100% in each case, as had been measured for the surface state of Gd(0001).^{19,20} Thus spin mixing within the exchange-split surface bands is expected to be negligible. This means that the occupied surface states are majority (spin-up) states, while the unoccupied surface states are minority (spin-down) states.

In Fig. 2, the lifetime widths Γ are plotted versus the energies E of the occupied and unoccupied surface states as listed in Table I. The results clearly show that for magnetic lanthanide metals, the lifetime widths of the majority states below E_F are considerably smaller than those of the minority states above E_F . This is also expressed by the two shaded areas in Fig. 2, light green (dark gray) for the occupied and light red (light gray) for the unoccupied surface states. In the following, we shall discuss various contributions to the linewidths, i.e., e -ph, e - e , e - m , and defect scattering.

Contributions by defect scattering can be excluded due to

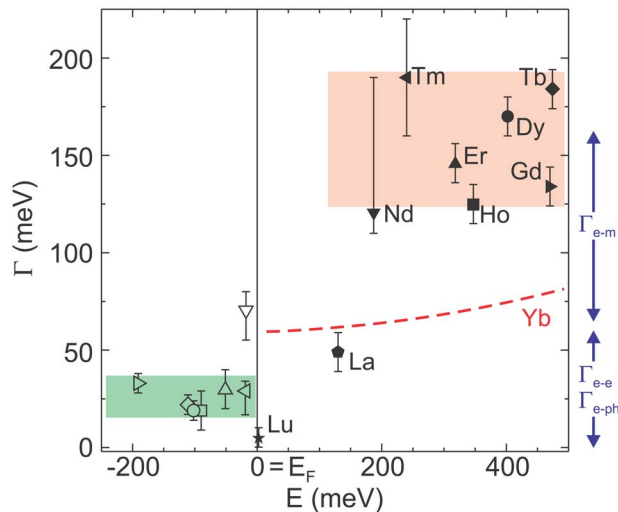


FIG. 2. (Color online) Lifetime width, Γ , vs energy, E , of occupied (open symbols) and unoccupied (filled symbols) surface states on trivalent lanthanide metals. The various lanthanide metals are distinguished by different symbols. The dashed red curve represents the lifetime widths of unoccupied quantum-well states in thin films of Yb metal (from Ref. 21). The vertical arrows on the right ordinate represent the estimated contributions of e -ph, e - e , and e - m scattering processes to Γ (for details, see text).

the cleanness of each studied surface that was verified locally prior to each STS measurement. Moreover, we checked the influence of rest-gas adsorbates and found that, in the case of the Dy(0001) surface, it influences the linewidth only within a radius of ≈ 20 Å. This observation actually reflects the high degree of lateral localization of these surface states.

Considering e -ph scattering, we note that—with the exception of Lu—the absolute values of the surface-state energies, $|E|$, are larger than the Debye energies of the respective lanthanide metals [$\hbar\omega_D < 20$ meV (Ref. 8)]. In these cases, the e -ph scattering contributions to Γ should be independent of energy.¹ Calculated values of $\Gamma_{e\text{-ph}}(T=0)$ range from 10 to 25 meV.⁸ Moreover, for the surface state on Gd(0001), the zero-temperature contribution of e -ph scattering to Γ has recently been determined as $\Gamma_{e\text{-ph}} = (22 \pm 5)$ meV.²² Thus, e -ph scattering accounts for $\approx 70\%$ of the total lifetime width Γ_{\uparrow} of the occupied surface state on Gd(0001), while it contributes to less than 20% of Γ_{\downarrow} . As similar values can be expected for the other lanthanide metals, we conclude that e -ph scattering cannot account for the large differences in Γ observed between the occupied and the unoccupied states.

According to three-dimensional Fermi-liquid theory, the e - e scattering rate is energy-dependent: $\Gamma_{e\text{-e}}(T=0) = 2\beta(E - E_F)^2$.²³ A fit of each data point in Fig. 2 by a parabola would lead to significantly different 2β values ranging from 0.4 to 1.4 eV⁻¹. These 2β values, however, are up to an order of magnitude larger than those reported for other materials.¹ In addition, we recently determined $2\beta \approx 0.05$ eV⁻¹ from the lifetime widths of quantum-well states (QWS) in thin Yb(111) films.²¹ Yb is a divalent lanthanide metal with $4f^{14}$ configuration. Although a comparison of QWS with surface states is not readily obvious, there

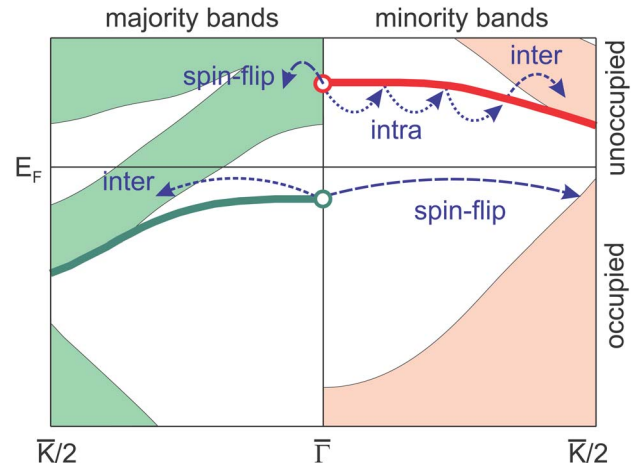


FIG. 3. (Color online) Schematic band-structure diagram of a magnetically ordered lanthanide metal showing decay channels for excited states close to $\bar{\Gamma}$, i.e., interband, intraband, and spin-flip transitions for an excited hole in the occupied (dark green line, left-hand side) and an excited electron in the unoccupied surface band (dark red line, right) at band maximum. The light green (dark gray) and red (light gray) areas mark majority and minority bulk bands projected to the (0001)-surface Brillouin zone, respectively (after Ref. 6). The lengths of the arrows reflect the momentum transfers Δk_{\parallel} needed for the indicated transitions.

is a close relationship between the electronic structure of QWS in the unoccupied Yb-bulk band and of the (0001)-surface states of trivalent lanthanide metals, in particular with respect to their k_{\parallel} dispersions.²⁴ Taking these facts into account, we conclude that large 2β values are not an adequate description of the situation.

A more specific view on the relaxation processes through e - e interactions is to consider intraband transitions within the surface band as well as interband transitions into bulk states, as shown schematically in Fig. 3. For surface states that lie in the center of a band gap of the surface-projected bulk band structure, interband-transition rates are expected to be low since such transitions involve large momentum transfers, Δk , and the Coulomb interaction scales with Δk^{-21} . Intraband transitions, on the other hand, may give significant, in some cases even dominant contributions to lifetime widths.²⁵ In the tunneling process, holes ($U < 0$) and electrons ($U > 0$) are predominantly excited at $\bar{\Gamma}$, i.e., at the maximum of the surface-state band. For occupied surface states, there are no intraband decay channels available for a hole at $\bar{\Gamma}$. Consequently, relaxation of the hole toward E_F can only take place via interband transitions, with the consequence that the overall contribution of e - e scattering in the occupied states is expected to be rather small. We therefore conclude that the dominant contribution to the lifetime widths of occupied surface states originates from e -ph scattering. For Lu, the surface-state energy is smaller than the Debye energy so that even e -ph scattering is suppressed,^{5,13} explaining the extremely small Γ value found for this metal (see Fig. 2).

On the other hand, an excited electron in an unoccupied surface band can decay into lower-lying electronic states of the same band. Therefore, intraband e - e scattering will be much stronger for unoccupied surface states. At first glance,

one could think that this decay channel explains the increased widths of unoccupied surface states. In fact, the Γ value of the unoccupied surface state on La(0001) exceeds those of the occupied surface states on the other lanthanide metals by about 25 meV (i.e., $\approx 100\%$), a clear signature of enhanced e - e scattering. However, the even larger difference of ≈ 100 meV between Γ values of the unoccupied surface states on the other lanthanide metals and the Γ value for La(0001) cannot be explained that way; this also applies to the deviation of Γ of the occupied surface state on Nd(0001) (∇ in Fig. 2).

For a consistent description of the observed lifetime widths, we have to consider also e - m scattering of surface-state electrons due to exchange interaction with $4f$ spins in the magnetic lanthanide metals. We first discuss a possible influence on the lifetime widths of occupied surface states. Except for Nd, the surface states are highly spin-polarized at 10 K, i.e., the intraband e - m scattering contribution should be weak due to the lack of final states of opposite spin within the surface band. Previous studies of Gd(0001) based on spin-resolved PE found an e - m -scattering contribution of ≈ 14 meV to the lifetime width of the occupied majority state, but only when the state was more than 25 meV—the highest magnon energy—below the band maximum.^{19,20,26} Due to the small dispersions of the surface-state bands, however, most excited holes will be closer to the band maximum so that the net contributions of intraband e - m scattering processes to the lifetime width of occupied surface states should be negligible.²² Regarding interband e - m transitions, the same argument as given before for e - e scattering applies in the case of occupied surface states, since the majority band is located within the total spin-averaged band gap.⁶ This would require large momentum transfers for interband transitions with spin-flip, rendering e - m scattering contributions negligible.

On the other hand, the unoccupied minority surface-state band lies outside the band gap of the projected majority band structure (see Fig. 3).⁶ Due to strong overlap with majority bulk bands, spin-flip transitions by magnon emission are possible with small Δk , and the transition rates will be strongly enhanced in the unoccupied surface band of the magnetic lanthanide metals. This explains the relatively large Γ values of unoccupied surface states as compared to those of the occupied states on magnetic lanthanide metals as well as of the unoccupied surface state on nonmagnetic La(0001).

As a consequence of the discussed processes, we draw the conclusion that the differences in lifetime widths of the unoccupied surface states on magnetic lanthanide metals, on the one hand, and the lifetime width of the surface state on La(0001), on the other hand, are predominantly due to e - m scattering. Then the e - m contribution amounts to $\approx (100 \pm 25)$ meV, where the error bar reflects the scattering of data points for different metals (see Fig. 2). We note here that the dominance of the e - m contribution to Γ is not astonishing, since a few experiments on Gd have already demonstrated that e - m coupling is rather strong in this magnetic metal.^{11,27,28} However, the finding that e - m scattering is strongly suppressed in the case of occupied surface states is noteworthy. The explanation presented here—a lack of final

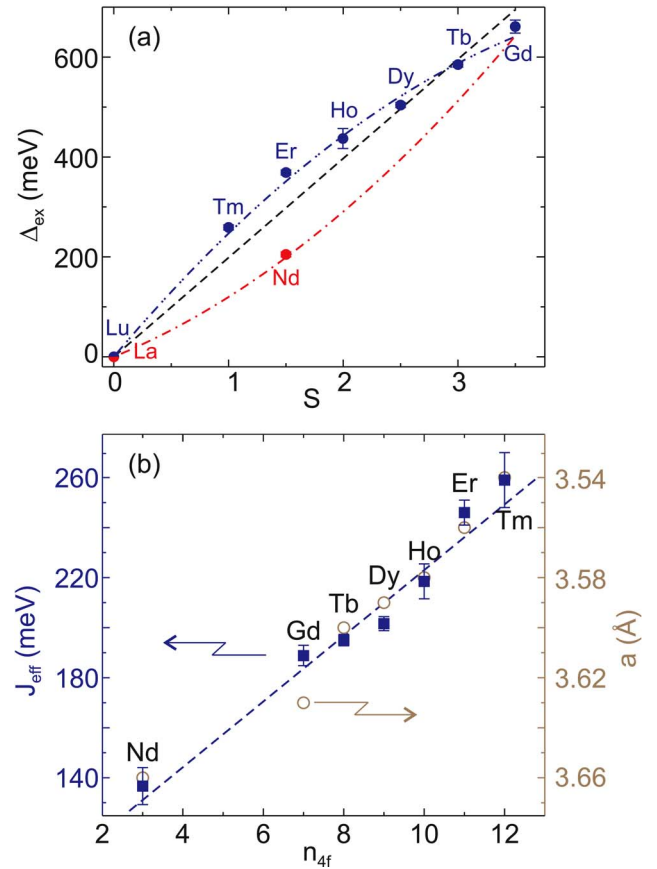


FIG. 4. (Color online) (a) Exchange splittings of (0001)-surface states, Δ_{ex} , at 10 K plotted vs $4f$ spins, S , of the studied lanthanide metals. Light lanthanide metals ($4f$ occupancy $n_{4f} < 7$) are plotted in red (light gray) circles, while the heavy ones ($n_{4f} \geq 7$) are displayed in blue (dark gray). The data points deviate significantly from the linear fit (black dashed line). (b) Effective exchange-coupling constant, J_{eff} , and lattice constant, a , vs n_{4f} of various lanthanide metals. In first approximation, $\Delta J_{\text{eff}} \propto n_{4f}$. This model describes the data for Δ_{ex} in (a) very well, both for the light (red dash-dotted curve) and the heavy lanthanide metals (blue dash-double-dotted curve); for details, see text.

states for spin-flip processes—is quite evident.

We further note that the deviation of the occupied surface state on Nd toward larger lifetime width can also be explained by e - m scattering. While the exchange-split surface states of the heavy lanthanide metals are almost 100% spin polarized, the net polarizations of the Nd surface states vanish due to the antiferromagnetic spin order within the (0001)-surface plane. This allows for enhanced intraband spin-flip scattering.

In the following, we shall discuss the systematic results of this study on the exchange splittings, $\Delta_{\text{ex}} = E_{\downarrow} - E_{\uparrow}$, which are plotted in Fig. 4(a) versus the $4f$ -spin quantum number S . While in a previous study $\Delta_{\text{ex}} \propto S$ was reported,⁵ a small but systematic deviation from a linear dependence is clearly observed in the present work. Furthermore, Nd exhibits a much smaller exchange splitting than Er, even though both elements have the same $4f$ spin. These findings are not in contradiction to the previous work, because there the number of data points had been too small to recognize these relatively

small but systematic deviations.⁵ Earlier PE experiments on bulk and interface states of the lanthanide metals were also unable to resolve such small variations within the limits of error.^{4,12,29} Instead, the present data unambiguously prove a nonlinear relationship between Δ_{ex} and S .

We introduce an effective exchange-coupling constant, $J_{\text{eff}} = \Delta_{\text{ex}}/S$, and plot in Fig. 4(b) the resulting J_{eff} versus n_{4f} . In this way, we find that J_{eff} is not constant for the series of lanthanide metals, but increases almost linearly with n_{4f} by $\approx 90\%$ from Nd to Tm. Another parameter that is known to change almost linearly with n_{4f} as a consequence of lanthanide contraction is the lattice constant a [open symbols in Fig. 4(b)]. This suggests a relationship between J_{eff} and a of the form $\Delta J_{\text{eff}} \propto -\Delta a$. Along these lines, the smaller exchange splitting observed for Nd as compared to Er—with both elements having identical $4f$ spin moments—can be readily understood. It is interesting to note that previous theoretical studies claimed a decrease of the atomic exchange-coupling constant, J , across the lanthanide series.^{18,30} In a crystal, however, the exchange splitting of a band scales with the ratio J/W , where W is the width of the band.³¹ Recent studies show that for the surface-state bands, W decreases across the lanthanide series,¹³ and this decrease is obviously larger than the decrease of J . The higher degree of localization (i.e., smaller bandwidth) of the $5d_{z^2}$ -surface states of the heavier

lanthanides is a consequence of lanthanide contraction, since the $5d$ states are contracted stronger than the delocalized $6s$ states. This explains why ΔJ_{eff} scales with $-\Delta a$.

With this model, the data for Δ_{ex} are well described for the light ($n_{4f} < 7$) and the heavy ($n_{4f} \geq 7$) lanthanide metals. For the light (heavy) lanthanide metals, one obtains $S = n_{4f}/2$ ($S = 7 - n_{4f}/2$). With $\Delta J_{\text{eff}} \propto n_{4f}$, this leads to $\Delta_{\text{ex}} \propto cS + S^2$ for light lanthanide metals [red dash-dotted curve in Fig. 4(a)], and to $\Delta_{\text{ex}} \propto (c+7)S - S^2$ for heavy lanthanide metals (blue dash-double-dotted curve); c is a common constant for the two regions.

In summary, the present work provides detailed information on the electronic structure of surface states on lanthanide metals, in particular on their energies, exchange splittings, and dynamics. We obtain a consistent description of the influence of lanthanide contraction on exchange splitting as well as the contributions of e -ph, interband and intraband e - e , and e - m scattering to the lifetime widths of the surface states, in the occupied and unoccupied regions around E_F .

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