Temperature dependence of lifetimes of Gd(0001) surface states

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Scanning tunneling spectroscopy was applied to study the temperature dependence of lifetime broadening of the Gd(0001) surface state. The observed increase of the linewidth with temperature is attributed to enhanced electron-phonon scattering, which can be described within the Debye model.

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Recently, the application of high-resolution photoelectron spectroscopy (PES),[1–3] scanning tunneling spectroscopy (STS),[4–6] and two-photon photoelectron spectroscopy (2PPES) (Refs. 7 and 8) for studying the electronic structure of metals and metal surfaces has led to considerable progress in understanding the dynamics of excited holes and hot electrons in these materials.[9,10] While 2PPES probes more or less directly the lifetime of electronic states, PES and STS measure the inverse of it, i.e., the lifetime broadening of the binding energies. Advantages of STS over PES consist in a higher energy resolution and in the ability to measure the density of states on both sides of the Fermi level, i.e., the density of occupied and unoccupied states. Major drawbacks of STS, on the other hand, are related to the strong influence of the electronic structure and geometry of the probing scanning-tunneling-microscope (STM) tip on the STS spectra which are generally not well known. In particular, if the density of states (DOS) of the tip is not structureless within the energy range of interest, all its features will appear in the STS spectra, convoluted with the electronic structure of the sample. Moreover, STS probes only states within some not well known fraction of the surface Brillouin zone, in general states around the $\bar{\Gamma}$ point.[11]

Here we use STS at variable temperature (10–100 K) in order to study the lifetime broadening of Gd(0001) surface states. Due to the high degree of three-dimensional localization of the Gd(0001) surface states, resulting in a rather small dispersion, the inherent restrictions of STS mentioned above do not apply here: The high surface-state DOS leads to distinct peaks in the tunneling spectra that can well be distinguished from tip induced features, and due to the almost negligible surface-state dispersion the exact knowledge of the *k*-space fraction probed by the STM tip is not of crucial importance. By analyzing our STS data, special attention was given to the contribution from temperature-dependent electron-phonon scattering. It turns out that the lifetime broadening of the surface-state peaks can be described within the Debye model with reasonable parameters for the electron-phonon mass-enhancement factor and Debye frequency.

The experiments were performed *in situ* in an ultrahigh vacuum system equipped with a low-temperature STM and sample-preparation facilities. The base pressure in the UHV system is typically 5×10^{-11} mbar. The STM consists of a commercial STM head (Omicron) mounted into a home-built liquid helium bath cryostat that allows for temperature control.[12] For STS measurements, the tunneling current I and the differential conductivity dI/dU are recorded as a function of sample bias at fixed STM-tip position (with a sweep rate of $\leqslant$100 mV/s). Close to the Fermi energy (E_F) dI/dU is to good approximation proportional to the local density of states of the sample at the energy $E-E_F=eU$. We measure the conductivity by modulating the bias voltage ($f_{\mathrm{mod}}\approx300$ Hz, $\Delta U^{\mathrm{r.m.s.}}\approx1$ mV) and detecting the amplitude of the tunneling-current modulation with a lock-in amplifier (time constant 100 ms).

Gd(0001) films of various thicknesses were deposited at room temperature on a clean W(110)-crystal surface by electron-beam evaporation out of a Ta crucible. The film thickness was controlled by a quartz microbalance with an error of $\pm20\%$. After deposition, the films were annealed at temperatures up to 1000 K controlled by a pyrometer. The quality and morphology of the films was checked by low-energy electron diffraction (LEED) and STM. For cleaning the W(110) surface and removing tungsten oxides, the crystal was annealed to a temperature above 2300 K. A detailed description of the sample preparation is given elsewhere.[12]

The tunneling spectra were taken on atomically flat terraces of the Gd(0001) films. The terraces were at least 10 nm wide so that due to the large effective mass of the surface states [$m^*/m>5$ (Ref. 12)] the impact of lateral confinement on the surface states should be negligible.[13] While the thicker films under investigation were flat and continuous, the thinner films [average thickness<3 monolayers (ML)] broke up into islands upon annealing (diameter>10 nm) with thicknesses ranging from 3 to 5 ML.

Figures 1(a)–1(d) show representative tunneling spectra obtained with a 35-ML-thick Gd/W(110) film at two different temperatures. Figure 1 shows typical STM images of this thick film with wide terraces (e) as well as of a 2.2-ML-thick Gd/W(110) film that broke up to 3-, 4-, and 5-ML-thick islands that are interconnected by a continuous Gd monolayer (f). The two peaks in the spectra in Figs. 1(a), 1(b) and 1(c), 1(d) correspond to the $5d_{z^2}$-like surface state, which is split due to exchange interaction with the $4f$ electrons to a majority-spin state below E_F (peak at $U\cong-180$ mV) and a minority-spin state above E_F (peak at $U\cong+470$ mV). The peaks could be fitted by Lorentzian functions convoluted with the derivative of the Fermi-distribution function. The latter has a width of about $3.5k_BT$ and defines the lower limit of experimental resolution. In principle, as was shown recently,[12] the peak shape is more complicated, exhibiting a small asymmetry due to a finite dispersion of the surface states. However, significant contributions to the peaks origi-

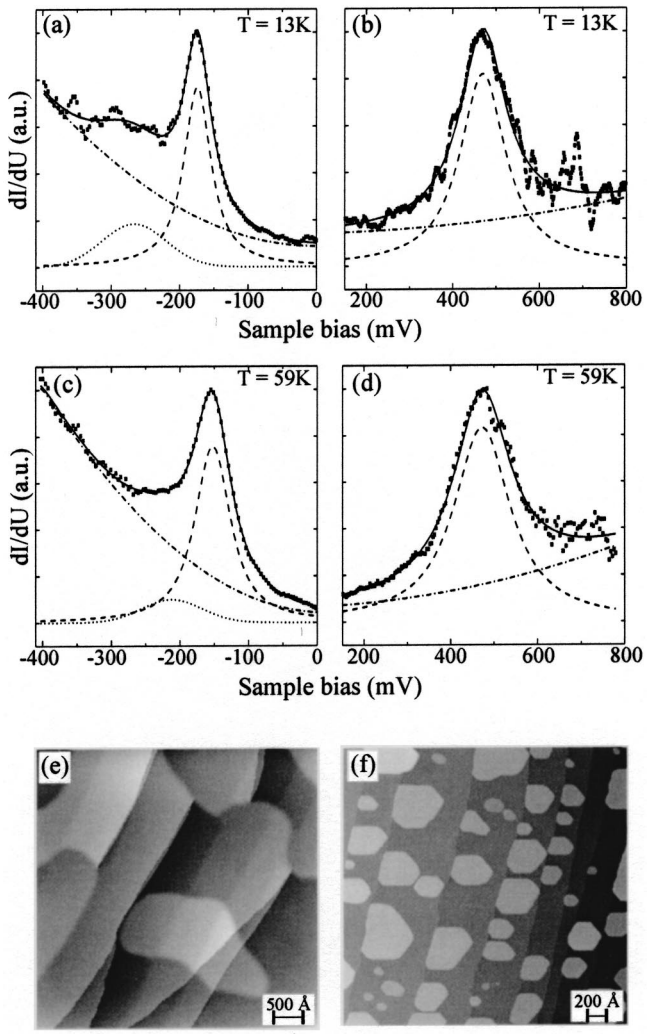

FIG. 1. Representative tunneling spectra recorded at two different temperatures, showing the exchange-split Gd(0001)-surface-state peaks below the Fermi energy (a), (c) and above (b), (d). Solid lines: Least-squares fits to the experimental data composed of a thermally broadened Lorentzian line (dashed), a quadratic background (dash-dotted), and a Gaussian line (dotted) that can be attributed to tip states (Ref. 12). STM images of a 35-ML-thick (e) and a 2.2-ML-thick (f) Gd/W(110) film annealed for about 5 min at 1000 and 870 K, respectively. The islands seen in (f) have thicknesses ranging from 3 to 5 ML.

nate from states within a binding-energy range of less than 15 meV below the band maximum at $\bar{\Gamma}$.[12] Since the exact degree of asymmetry depends apparently on the geometry or electronic structure of the STM tip, and since the error of the Lorentzian linewidths by neglecting the asymmetry effect is relatively small (<10 meV), the peaks in the spectra were fitted just with a single Lorentzian line.

Between 10 and 100 K, the energy position of both the majority and the minority surface-state peak shifts by less than $\Delta E = 20$ meV (the energy is given by $E = eU$). The width of the peaks, on the other hand, increases with increasing temperature. Since the Lorentzian linewidth Γ is related to the lifetime τ of the excited state by $\Gamma = \hbar/\tau$, it means that the lifetime of excited holes (below E_F) or electrons (above E_F) decreases with temperature. In Figs. 2(a) and 2(b) the linewidths are plotted as a function of temperature for the occupied and the unoccupied surface state, respectively. The data points and estimated error bars result from averaging the linewidths of only the best spectra with lowest noise out of numerous sets of spectra recorded at different positions on the samples.

The lifetime of an excited localized state is determined by the electron-electron (*e-e*) (Ref. 14) and electron-phonon (*e-*ph) scattering rate[2,15,16] and—in the case of magnetic materials—by an additional electron-magnon (*e-m*) interaction.[17] As we shall discuss below, the *e-m* scattering contribution to the linewidth of a surface-state peak in the tunneling spectra is assumed to be small and is neglected here. Concerning the temperature dependence of the lifetime broadening, it is mainly given by the temperature dependence of the *e*-ph scattering rate (see next paragraph). The *e-e* scattering rate, on the other hand, shows only a weak temperature dependence with changes $\Delta\Gamma_{e\text{-}e}$ of less than 1 meV for $T < 100$ K.[2,12,14] An implicit temperature dependence of $\Gamma_{e\text{-}e}$ may arise from the rather strong energy dependence of $\Gamma_{e\text{-}e}$ as well as a temperature-dependent surface-state binding energy. However, for $T < 100$ K, the observed energy shifts (resulting from a temperature dependence of the exchange splitting of the surface state) are less than 20 meV, which would lead to $\Delta\Gamma_{e\text{-}e} < 4$ meV for the occupied surface state (binding energy $E \cong -180$ meV) and $\Delta\Gamma_{e\text{-}e} < 8$ meV for the unoccupied surface state ($E \cong 470$ meV).[12] These changes are within the error bars and can be neglected in the present context.

Within the Debye model, the *e*-ph scattering contribution to the linewidth $\Gamma_{e\text{-ph}}$ as a function of binding energy E and temperature T is given by[2,15]

$$\Gamma_{e\text{-ph}}(E,T) = \lambda \frac{2\pi}{(\hbar\omega_D)^2} \int_0^{\hbar\omega_D} dE' E'^2 [1 - f(E-E') + 2n(E') + f(E+E')], \quad (1)$$

with ω_D being the Debye frequency, λ the electron-phonon mass-enhancement factor, $f(E)$ the Fermi distribution, and $n(E)$ the Bose-Einstein distribution. The total linewidth Γ is given by

$$\Gamma(E,T) = \Gamma_{e\text{-ph}}(E,T) + \Gamma_{e\text{-}e}(E). \quad (2)$$

We point out that Eq. (1) is valid, although it is derived from a three-dimensional Debye model, because for small energies, the two-dimensional Debye model would approximately lead to the same result.[16] At high temperatures, $\Gamma_{e\text{-ph}}$ shows a linear temperature dependence $d\Gamma_{e\text{-ph}}/dT \cong 2\pi\lambda k_B$ (for $k_B T > \hbar\omega_D$ and $|E - E_F| > \hbar\omega_D$), i.e., independent of ω_D. Recent PES measurements of the occupied Gd surface state for temperatures up to 160 K (Ref. 3) and 400 K (Ref. 18) confirm a linear temperature dependence of $\Gamma_{e\text{-ph}}$. From the temperature gradients, an electron-phonon mass-enhancement factor $\lambda = 1.0 \pm 0.2$ is deduced. This value is compatible with the result of specific-heat measurements,[19] however, it is significantly larger than a calculated value ($\lambda_{th} = 0.4$).[19]

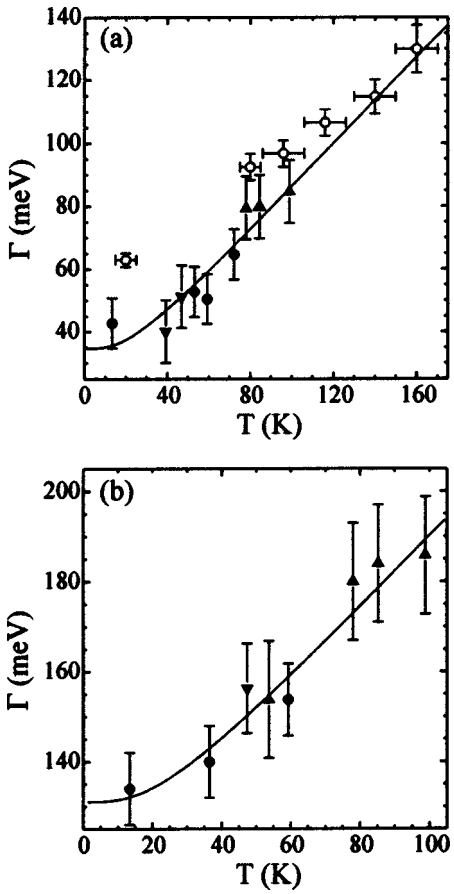

FIG. 2. Lifetime broadening (FWHM) of the occupied (a) and the unoccupied (b) Gd-surface state as a function of temperature as obtained from the present STS data (solid symbols) and the PES study by Fedorov *et al.* (Ref. 3) (open circles). The STS data was obtained with a 35-ML-thick Gd/W(110) film (●) as well as with 4-ML- (▲) and 5-ML- (▼) thick Gd islands. In general, data recording became less stable at elevated temperatures (mostly due to temperature drift). In particular, instabilities were observed for the thin Gd/W(110) films, which might have caused some peak broadening in the tunneling spectra recorded on the Gd islands. The curves (solid lines) in the two diagrams have been calculated using Eq. (1) for $\hbar\omega_D = 8$ meV and $\lambda = 1.3$ (a) and $\lambda = 1.5$ (b), respectively.

We have recorded tunneling spectra only at low temperatures ($T < 100$ K), where the temperature dependence of $\Gamma_{e\text{-ph}}$ deviates from a linear function. A quantitative description based on Eqs. (1) and (2) requires the knowledge of three parameters λ, ω_D^{surf}, and $\Gamma_{e\text{-}e}$. There are not enough data points and sufficiently small error bars to allow for an unambiguous determination of all three parameters by least-squares fitting. Therefore, we set ω_D^{surf} to a physically reasonable value and determined the other two parameters. Alternatively, we could have used the value of λ recently determined by PES and use ω_D^{surf} as a fit parameter. However, since a much smaller value for λ was found by theory,[19] we pursued the other approach to see whether the experimental or the theoretical result could be confirmed: The Debye frequency for bulk Gd is $\hbar\omega_D^{\text{bulk}}$ (Gd) = 14 meV.[20] At the surface, the Debye frequency is likely to be reduced by up to a factor of 2 with respect to the bulk value.[21] Surface-Debye frequencies have been determined for the lanthanide metals Tb, Yb, and Lu. In all cases, $\omega_D^{\text{surf}} = (0.6 \pm 0.1)\omega_D^{\text{bulk}}$.[22] Because of the similar chemical and electronic properties of the lanthanides, we assume that the surface Debye frequency of Gd is reduced to 60% of its bulk value as well.

By fitting our data (Fig. 2) with $\hbar\omega_D^{\text{surf}} = 8$ meV ($\cong 0.6\hbar\omega_D^{\text{bulk}}$), we obtain for the occupied surface state $\lambda = 1.3 \pm 0.3$ and $\Gamma_{e\text{-}e} = 13 \pm 10$ meV. For the unoccupied surface state, we find $\lambda = 1.5 \pm 0.3$ and $\Gamma_{e\text{-}e} = 106 \pm 10$ meV (see curves in Fig. 2). Within the error bars, the values for λ agree with the value found by PES (for the occupied Gd surface state),[3,18] which clearly shows that the electron-phonon coupling strength has been underestimated in previous calculations.[19] It should be noted that fits of the STS data for larger (smaller) surface Debye frequencies resulted in larger (smaller) values for λ. For example, if we would ignore a reduction of the Debye frequency at the surface, the values for λ were about 30% larger and no longer compatible with the PES results.

For comparison of our data with results of the spin-resolved PES study by Fedorov *et al.*[3] we have plotted their data in Fig. 2(a). The data points correspond to the Lorentzian linewidths of the majority-spin component of the occupied surface-state peak in the photoemission spectra (there is also a finite minority-spin component, which contributes 13% to the spin-integrated PES peak[3]). Compared to the STS data we see that at low temperatures the linewidths derived from the PES data are by about 20 meV larger than those of the STS peaks.

Provided that in both cases the experimental resolution and possible artifacts have been accounted for in a proper way, a plausible explanation for this discrepancy is that PES measures the average over a relatively large surface area, i.e., lateral inhomogeneities of the surface-state binding energy would lead to a broadening of the peaks. In fact, we have observed position-dependent variations of the binding energy on the same Gd film by more than ± 10 meV.

At this point, we want to comment on the suggestion that electron-magnon scattering gives a significant contribution to the lifetime broadening of the surface-state peaks in the photoemission spectra. For the majority-spin component of the occupied surface state it was estimated to amount to $\cong 14$ meV.[3,17] However, the *e-m* scattering rate depends on the surface-state energy with respect to the maximum of the surface-state band, and an inverse lifetime of 14 meV is only expected for energies beyond 25 meV. For smaller energies, the *e-m* scattering rate is reduced, and it vanishes at the band maximum. As mentioned above, the energy range below the maximum of the surface-state bands that contributes to the peaks in the tunneling spectra is supposed to be narrower than 15 meV.[12] Therefore, we would expect a strongly reduced contribution of *e-m* scattering to the surface-state lifetimes. The reason for this narrow energy range is a very high effective mass, i.e., a high degree of localization of the surface states. The STS results indicate that the effective masses of the Gd(0001) surface states are significantly larger than the value obtained by Fedorov *et al.*, $m^*/m = 1.21$:[3,17] For the sp-like surface states in Cu(111), Ag(111), and Au(111),

with well-known effective masses of $m^*/m(\mathrm{Cu,Ag}) \cong 0.4$ and $m^*/m(\mathrm{Au}) \cong 0.3$,[10] almost steplike functions were measured by STS.[4,23] Since we find sharp peaks with very small asymmetry, we can find a lower limit for the effective masses of the Gd(0001) surface states of $m^*/m(\mathrm{Gd}) \geqslant 5$. By inspection of the data in Ref. 3, it is likely, however, that the effective mass is a function of energy and decreases as the surface-state band approaches the edges of the band gap in the projected band structure.[24]

Finally, we want to comment on the *e-e* scattering contribution to the surface-state linewidths, $\Gamma_{e\text{-}e}$. Within Fermi-liquid theory,[14] $\Gamma_{e\text{-}e}$ is approximately given by $\Gamma_{e\text{-}e} \cong 2\beta(E - E_F)^2$. With $\Gamma_{e\text{-}e}(-180\ \mathrm{meV}) = 13 \pm 10\ \mathrm{meV}$ and $\Gamma_{e\text{-}e}(470\ \mathrm{meV}) = 106 \pm 10\ \mathrm{meV}$, the factor β—describing the strength of the *e-e* interaction—amounts to $\beta = (0.20 \pm 0.15)\ \mathrm{eV}^{-1}$ and $(0.24 \pm 0.03)\ \mathrm{eV}^{-1}$ for the occupied and unoccupied surface state, respectively. We point out that these values differ slightly from those deduced in our previous work, where we assumed a smaller theoretical value for λ and used the bulk Debye frequency.[12]

In summary, we have investigated the temperature dependence of the surface-state lifetime for the Gd(0001) surface by STS. The observed increase of lifetime broadening of the surface-state peaks in tunneling spectra with increasing temperature could be described within the Debye model for electron-phonon scattering. By using a reduced surface Debye energy, $\hbar\omega_D^{\mathrm{surf}} = 8\ \mathrm{meV}$, good fits to the data could be obtained with an electron-phonon mass-enhancement factor $\lambda = 1.3 \pm 0.3$ ($\lambda = 1.5 \pm 0.3$) for the occupied (unoccupied) surface-state peak, which is consistent with previous experimental results.

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