Spin-resolved photoemission study of photohole lifetimes in ferromagnetic gadolinium

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High-resolution spin-resolved photoemission is used to probe the decay channels available to a photohole created in a Gd(0001) surface state. The photoemission linewidths show that at low temperatures, the lifetime of a majority-spin hole is predominantly limited by electron-phonon scattering and that of a minority-spin hole by electron-magnon scattering. With increasing temperature this state shows both spin-mixing behavior reflecting the exchange of magnons and a reduced exchange splitting.

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In photoemission a photon of known energy ω is adsorbed and the outgoing electron's energy $(\omega-\varepsilon_k)$ and momentum are measured.[1] Interaction effects cause the sharp line spectrum with shape defined by $\mathrm{Im}\,1/(\omega-\varepsilon_k-i\eta)$ arising from independent electron theory to evolve into $\mathrm{Im}\,1/[\omega-\varepsilon_k-\Sigma(k,\omega)]$ where the complex self-energy $\Sigma(k,\omega)$ contains the effects of the interactions. The real part $\Sigma_1(k,\omega)\sim-\omega\lambda_k$ gives a shift in energy and mass enhancement, while the imaginary part $2\Sigma_2(k,\omega)=\Gamma_k(\omega,T)$ gives the lifetime broadening $\hbar/\tau_k$ where τ_k is the lifetime. Recent improvements in the energy and momentum resolution of photoemission have enabled detailed studies of self-energy effects reflecting various interactions including the electron-phonon coupling.[2,3] In a study of Mo, contributions to $\Gamma_k(\omega,T)$ reflecting electron-phonon, electron-electron, and electron-impurity scattering were all resolved.[2] These studies have led to an intense discussion in the field of high T_c superconductivity where a mass renormalization in the vicinity of the Fermi level has been attributed alternatively to coupling to phonons[4] and to spin excitations.[5] The cuprates represent doped antiferromagnetic insulators. Here, we report a spin-resolved study of lifetime effects in a localized ferromagnetic system, namely Gd. In such a system the possibility again exists for a hole to scatter from spin excitations as well as phonons. Surprisingly, we demonstrate that at low temperatures, a majority-spin photohole relaxes mainly by phonon emission whereas a minority-spin hole relaxes mainly by spin-wave emission. A preliminary report of some aspects of this work has been given elsewhere.[6]

The experiments employed a Scienta SES200 electron energy analyzer,[6] which can be used either for high-resolution angle-resolved photoemission or for spin-resolved photoemission. Spin polarization is detected with a micro-Mott polarimeter, modified from the design of the Rice University Group.[7] UV photons of energy 21.2 eV were provided by a resonance lamp. The energy resolution in both the angle-resolved and spin-resolved studies is ~50 meV. An angular resolution of $\Delta\theta\sim0.2°$ is achieved without spin resolution but for spin-resolved studies, because of the reduced intensity, $\Delta\theta\sim\pm3°$. Gadolinium films of thickness 200 Å were evaporated onto a Mo(110) substrate at room temperature and annealed to 750 K to produce well-defined surfaces with a single magnetic domain.[8] Low-energy electron diffraction (LEED) monitored the crystallographic order.

Figure 1 shows spectral density maps in the ΓX azimuth from the clean Gd(0001) surface at different temperatures. The width, $\hbar/\tau=-2\,\mathrm{Im}\,\Sigma$, of the surface state at a binding energy of ~170 meV increases as the temperature is raised from 82 to 300 K indicating a reduction in the lifetime as a result of increased electron-phonon and electron-magnon scattering at higher *T*. In the low-temperature plot, the surface state has $\hbar/\tau\sim$const until the emission angle exceeds 5°. At this point, band-structure calculations[9] indicate that the surface state leaves the bulk band gap and begins to resonate with bulk bands.

Figure 1 also shows a temperature-dependent shift of 75 meV in the binding energy of the state. Nolting *et al.*[10] have shown that, depending on the coupling between conduction and localized electrons, the quasiparticle band may show either a Stoner-like reduction of exchange splitting with increasing temperature (weak coupling) or temperature-dependent spin mixing without reduction of exchange splitting (strong coupling). At intermediate coupling, a mixture of the two behaviors is anticipated. Fedorov *et al.*[11] and Weschke *et al.*[12] both measured a reduction of the exchange splitting with increasing temperature. Similar behavior was seen in a spin-polarized inverse photoemission by Donath *et al.*[13] In contrast, Li *et al.*,[14] using spin-polarized photoemission, saw a spin-mixing behavior. In a recent scanning tunneling spectroscopy (STS) study, Getzlaff *et al.* saw a mixture of the two behaviors.[15] With higher energy resolution than previous spin-resolved photoemission studies[14] we confirm the results found in the STS study.[15]

Figure 2 shows spin-polarized photoemission spectra recorded from the surface held at $T=20$ K in an angular acceptance $\Delta\theta\sim\pm3°$. Such an angular acceptance adds only minimally to the measured linewidths. The peaks in the majority- and minority-spin spectra occur at nearly identical binding energies. Earlier experiments[16,17] and calculations[18] indicate that the surface state should be 100% majority spin, due to parallel alignment of the surface and bulk moments. Our observation of a minority-spin component could be taken as evidence of either incomplete saturation of the magnetization or the presence of minority domains. However, the

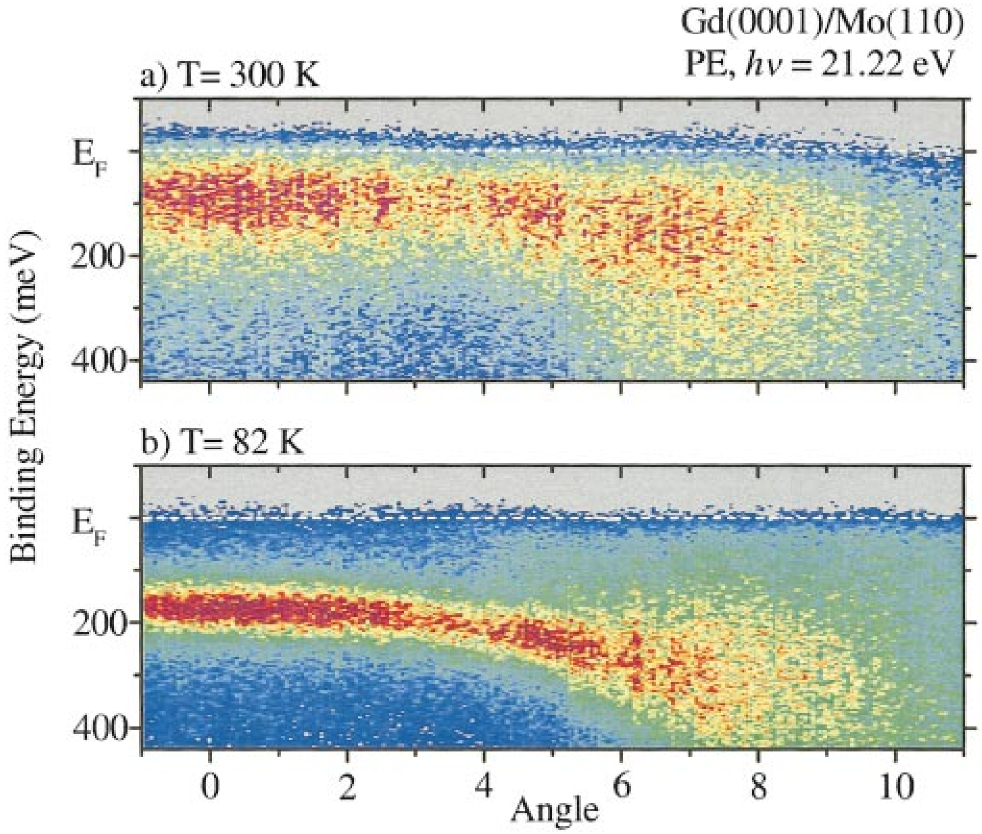

FIG. 1. (Color) Upper panel: Spin-integrated photoemitted spectral response for the Gd(0001) surface as a function of binding energy and angle of emission measured from the surface normal. The sample T is 300 K and the incident photon energy is 21.2 eV. Lower panel: As above but now the sample T is 82 K.

growth procedure is known to yield a single magnetic domain.[8] The coexistence of both spin components at the same energy is therefore an intrinsic property of the surface state arising from a combination of spin-orbit and spin-exchange processes as discussed by Nolting *et al.*[10] A simple model allows us to quantify the effect of the former giving a polarization $P=\Delta/\sqrt{(\Delta^2+\zeta^2)}$ in each quasiparticle state. With a spin-orbit parameter $\xi=0.3$ eV and exchange splitting $\Delta=0.7$ eV at 0 K, we get a spin-orbit induced mixing $R=(n_\downarrow/n_\uparrow)=(1-P)/(1+P)\sim 5\%$. This increases to 8% at $T=150$ K because of the reduced exchange splitting.

Fitting the spectra in Fig. 2 with Lorentzians shows that the minority-spin peak has a larger width than its majority-spin counterpart, 116 meV as opposed to 86 meV. Extracting the experimental resolution these widths become approximately 105 meV in the minority-spin channel and 70 meV in the majority channel. The lifetime of a photohole can be limited by electron-electron, electron-phonon, and electron-impurity scattering, and in a magnetic system, by electron-magnon scattering. At the edge of the surface band, the state also couples to the bulk states as seen in Fig. 1 for $\theta > 5°$. However, such coupling will be stronger in the majority-spin channel since the state falls closer to the bulk majority-spin band edge than to the minority-spin counterpart.[9]

Each scattering mechanism gives distinct spin-dependent contributions to the total scattering rate. Electron-electron scattering by exchange processes favors the two holes in the final state being of opposite spin.[19] From consideration of the spin-dependent densities of states, we estimate the scattering rate from this process to be equal for majority- and minority-spin holes. The electron-phonon and impurity scattering rate are proportional to the density of states at the hole binding energy for the same spin while the electron-magnon rate is proportional to the density of states for the opposite spin. Since the majority-spin density of states is large and the minority-spin component small, impurity and electron-phonon scattering should be most important in the majority-spin channel. The fact that the minority-spin channel is broader indicates an electron-magnon mechanism. At $T=0$ K, the minority-spin component of the photohole can scatter to the majority-spin component higher in the surface band by emitting a spin wave. The corresponding spin-flip process is not available to the majority-spin component of the photohole at $T=0$ because the localized f spins have saturated magnetization. At higher temperatures, inelastic scattering can occur back and forth between the two spin channels mediated by the emission or absorption of magnons, but the minority-spin component always has the higher density of final states to scatter into. An approximate treatment[20] using the "s-f" Hamiltonian[21] finds the result

$$\hbar/\tau(\downarrow)=\frac{\sqrt{3}}{4}\,\frac{P'(\uparrow)m^*}{S}\left(\frac{2JSa}{\hbar}\right)^2 \tag{1}$$

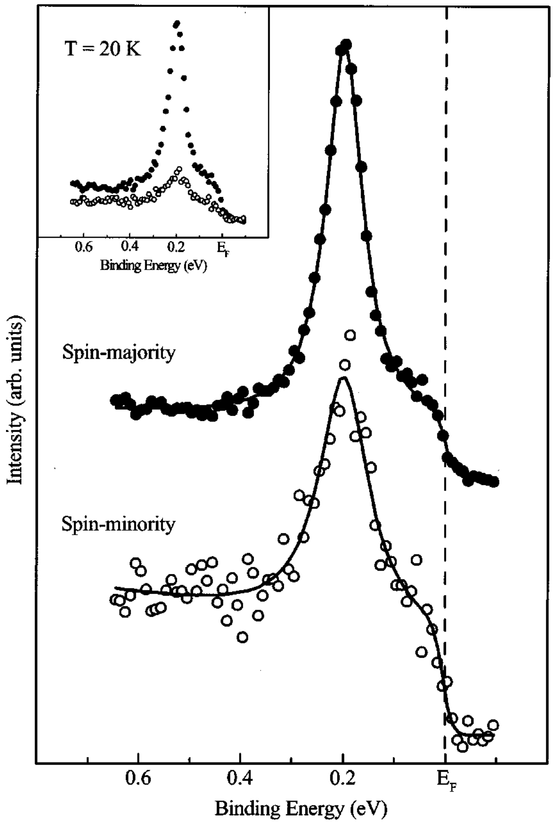

FIG. 2. Spin-resolved photoemission spectra recorded from the Gd(0001) surface at 20 K. The upper and lower spectra represent the emission in the majority- and minority-spin channels, respectively. The lines indicate Lorentzian fits to the spectra superimposed on appropriate backgrounds. The inset shows the relative intensities in the two spin channels.

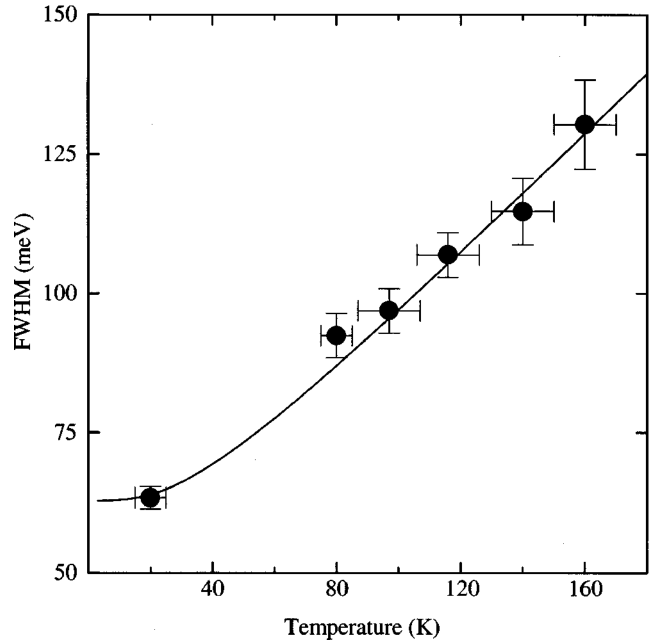

FIG. 3. The full width half maximum (FWHM) of the majority-spin peak as a function of T. The solid line indicates a fit to the data using Eq. (2) as given in the text.

for the decay of the minority (↓) spin component due to spin-flip scattering with magnon emission. Here J is the s-f exchange parameter giving the exchange splitting $2JS = 0.65$ measured[9] for the surface state, $m^* = 1.21$ is the effective mass measured for the surface band, and $P'(\uparrow) = 0.87$ is the experimentally measured majority component of the band. With $S = \frac{7}{2}$ and $a = 3.6$ Å, $\hbar/\tau(\downarrow) \approx 0.095$ eV. Conversely, replacement of $P'(\uparrow)$ by $P'(\downarrow) = 1 - P'(\uparrow)$ gives $\hbar/\tau(\uparrow) \approx 0.014$ eV for the majority-spin component. Thus at low T, the majority-spin channel is dominated by electron-phonon scattering whereas the minority-spin channel is dominated by electron-magnon scattering. Using the same densities of states arguments we can anticipate the opposite behavior in the unoccupied bands. Thus at low T, the decay of an excited electron in the unoccupied majority-spin band will preferentially involve magnons and the decay of an excited electron in the unoccupied minority-spin band will preferentially involve phonons.

Turning to the temperature dependence of the line shapes, first consider electron-electron scattering. As the hole moves to lower binding energies, the phase space for such scattering is reduced. On the contrary, Fig. 1 shows that as T is raised, the state moves closer to E_F and the peak broadens. This indicates that electron-phonon and electron-magnon scattering play the dominant role and not electron-electron scattering. Considering electron-phonon coupling alone, the width or inverse lifetime of the photohole is described by the relationship

$$\frac{\hbar}{\tau}(\omega,T) = 2\pi \int_0^\infty d\omega' \, \alpha^2 F(\omega')[1 - f(\omega - \omega') + f(\omega + \omega') + 2n(\omega')], \tag{2}$$

where $f(\omega)$ and $n(\omega)$ are Fermi and Bose distribution functions and $\alpha^2 F(\omega)$ is the Eliashberg coupling function. For $T \geqslant \Theta_D/3$ (Θ_D is the Debye temperature) the phonon contribution to the width is $\hbar/\tau = 2\pi\lambda k_B T$, that is, linear in T. As shown in Fig. 3, fitting the experimentally determined majority-spin linewidths with the expression given in Eq. (2) leads to a value of the electron-phonon coupling constant $\lambda \approx 1.0$ for the majority-spin channel. This value may be compared with a value of 1.2 (bulk, spin averaged), extracted from the measured specific heat[22] and a theoretical value of 0.4 (also bulk and spin averaged) obtained in a spin-polarized calculation of the electron-phonon coupling constant.[23]

The electron-phonon coupling parameter may be written as $\lambda = N_S \langle I_S^2 \rangle / M \langle \omega^2 \rangle$ where N_S represents the spin-projected density of states at the hole binding energy, $\langle I_S^2 \rangle$ is the Fermi surface average of the electron-phonon matrix element, M is the atomic mass, and $\langle \omega^2 \rangle$ is an average phonon frequency. Following the analysis of Skriver and Mertig,[23] but allowing for spin-dependent coupling, one arrives at values $\lambda \approx 0.73$ and 0.31 for the majority- and minority-spin bulk bands, respectively. Our value for the majority-spin channel is higher, reflecting perhaps a higher electron density of states in the surface region. Wu *et al.* have in fact calculated an enhanced magnetic moment in the surface layer.[9] Using their calculated majority- and minority-spin densities, one obtains $\lambda \approx 1.15$ and 0.25 for the surface majority-and minority-spin electron-phonon coupling. The spin-dependent density of states available for scattering is not constant but varies with T as shown in Fig. 1. A rough estimate suggests that λ could change by as much as 10% over the range of T in Fig. 3, decreasing for the majority-spin channel and increasing for the minority-spin channel. Electron-magnon scattering will show the opposite behavior, increasing with T in the majority-spin channel and decreasing in the minority-spin channel.

In summary, our spin-resolved photoemission studies with high-energy resolution have allowed us to directly measure the different contributions to the spin-dependent lifetimes in Gd. We show that at low T, the majority-spin lifetime is predominantly limited by electron-phonon scattering whereas the minority-spin lifetime is determined by electron-magnon scattering. As T increases there is an increasing admixture of the two decay modes in each channel. The observation of a magnon contribution to the decay of the excitations is consistent with de Haas–Van Alphen studies of cyclotron masses in gadolinium which suggests that electron-magnon scattering makes a significant contribution to the mass enhancement.[24]

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