Exact theory for electronic Raman scattering of correlated materials in infinite dimensions

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A wide variety of strongly correlated materials including SmB_6, FeSi, and the underdoped cuprates display anomalous behavior in their Raman response, which includes a low-temperature transfer of spectral weight from low to high energy (as T is reduced) and the appearance of an isosbestic point (a characteristic frequency where the Raman response is independent of temperature). We illustrate how these features appear in the Raman response of the infinite-dimensional Hubbard model, which is the simplest system to undergo the Mott transition from a Fermi liquid phase. We find that the qualitative behavior in the insulating phase is model independent, and that a number of new features arise as one approaches the metal-insulator transition from the Fermi-liquid phase. Such behavior has not yet been seen in experiment. We propose a number of different systems that are likely to show these new anomalies.

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Strongly correlated systems as disparate as mixed-valence compounds[1] (such as SmB_6), Kondo insulators[2] (such as FeSi), and the underdoped cuprate high-temperature superconductors,[3–5] show temperature-dependent B_{1g} Raman spectra that are both remarkably similar and quite anomalous (see Fig. 1). This experimental ''universality'' (or material independence) suggests that there is a common mechanism governing the electronic transport in correlated insulators. As these materials are cooled, they all show an increase in the spectral weight in a ''charge-transfer'' peak with a simultaneous reduction of low-frequency spectral weight. This spectral weight transfer is slow at high temperatures and then rapidly increases as the temperature is lowered in the proximity of a quantum-critical point (corresponding to a metal-insulator transition). The Raman spectral range is also separated into two regions: one where the response decreases as T is lowered and one where the response increases. The characteristic frequency that divides these two regions is called the isosbestic point, which is the frequency where the Raman response is independent of temperature. These anomalous features are not typically seen in either the A_{1g} or the B_{2g} channels.

This anomalous experimental behavior is plotted in Fig. 1. The top panel shows SmB_6, which has the added feature of developing a sharp peak at 130 cm^{-1} (which does not disperse in frequency) when the temperature is lower than 30 K. The FeSi data are shown in the middle panel. FeSi displays the cleanest signature of these anomalous features. Note how the isosbestic point only develops at temperatures below 150 K. The bottom panel shows smoothed data in the lanthanum strontium copper oxide high-temperature superconductor. The isosbestic point is somewhat harder to see here (because of the noise in the data), but it does develop at about 2100 cm^{-1} as the temperature is lowered.

The theoretical description of Raman scattering in correlated materials has been stymied by the lack of techniques that can accurately treat electron-electron interactions and incipient quantum phase transitions to evaluate the Raman response. An exact treatment of Raman scattering in weakly interacting metals is missing. It is well known that in the absence of inelastic scattering, the low-energy Raman cross section vanishes at $q=0$ due to the lack of phase space available to create particle-hole pairs. Therefore any signal at all must come from electron-electron interactions[6] or impurities.[7] Shastry and Shraiman[8] (SS) outlined a procedure to construct a theory that can interpolate from a metal to an

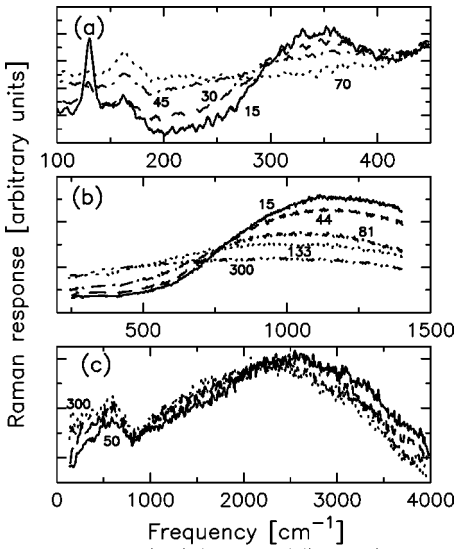

FIG. 1. Experimental Raman response for correlated materials (a) SmB_6, Ref. 1; (b) FeSi, Ref. 2; and (c) underdoped $La_{2-x}Sr_xCuO_4$, Ref. 3, with $x=0.08$. All of the experimental data show the development of a low-temperature isosbestic point, which occurs due to the transfer of spectral weight from low energy to high energy as the temperature is lowered, indicating the proximity to the quantum-critical point of a metal-insulator transition. The individual curves are labeled by the temperature in K where the measurement was taken. In panel (c) only the high-temperature (300 K) and the low-temperature (50 K) curves are labeled. The two intermediate curves are at 100 and 200 K, respectively.

insulator. However, due to the ever increasing Hilbert space needed to describe a metal from the insulating side, or the lack of a clear picture of quasiparticles from the metallic side, quantitative calculations were not feasible. One is then left with approximate methods or phenomenological approaches to construct a Raman theory that suffer the limitations of being unable to reach different phases and of lacking a microscopic basis.

An exact solution[9,10] on the simpler Falicov-Kimball model[11] produced the qualitative behavior seen in correlated insulators, and it was argued that Raman scattering results should be model independent in the insulating phase. But most real materials have an underlying Fermi-liquid phase close to the metal-insulator transition, and the Falicov-Kimball model is never a Fermi liquid, so it is important to investigate what happens in a true Mott transition. Indeed this is the point of this contribution. We find that a strongly correlated Fermi liquid has a number of striking features in its Raman response, and we suggest a number of candidate materials to observe this new behavior. Our analysis is restricted to the nonresonant response and the local approximation of dynamical mean field theory; hence we are unable to treat both resonant effects and interactions with nonlocal excitations (like spin waves). Nevertheless, one learns much from our restricted (but exact) treatment. The infinite-dimensional Hubbard model has already been investigated using perturbation theory[12] but not in the vicinity of the metal-insulator transition.

The nonresonant Raman scattering in the B_{2g} channel vanishes,[10] and in the A_{1g} channel it requires knowledge of the local irreducible charge vertex (which is problematic to calculate in the infinite-dimensional Hubbard model), so we provide results only for the B_{1g} sector here. The Hubbard Hamiltonian[13] contains two terms: the electrons can hop between nearest neighbors [with hopping integral $t^*/(2\sqrt{d})$ on a $(d\rightarrow\infty)$-dimensional hypercubic lattice[14]], and they interact via a screened Coulomb interaction U when they sit on the same site. All energies are measured in units of t^*. The Hamiltonian is

$$H=-\frac{t^*}{2\sqrt{d}}\sum_{\langle i,j\rangle,\sigma} c_{i\sigma}^{\dagger}c_{j\sigma}+U\sum_i n_{i\uparrow}n_{i\downarrow}, \qquad (1)$$

where $c_{i\sigma}^{\dagger}$ ($c_{i\sigma}$) is the creation (annihilation) operator for an electron at lattice site i with spin σ and $n_{i\sigma}=c_{i\sigma}^{\dagger}c_{i\sigma}$ is the electron number operator. We adjust a chemical potential μ to fix the average filling of the electrons.

The nonresonant Raman response in the B_{1g} channel has no vertex corrections[9,10,15] and is equal to the bare bubble. The derivation of the Raman response has been presented elsewhere[10] and can also be found from the SS relation[8,16]—the imaginary part of the nonresonant B_{1g} Raman response is

$$\mathrm{Im}R(\nu)=c\int d\omega[f(\omega)-f(\omega+\nu)] \times\int d\epsilon\,\rho(\epsilon)A(\epsilon,\omega)A(\epsilon,\omega+\nu), \qquad (2)$$

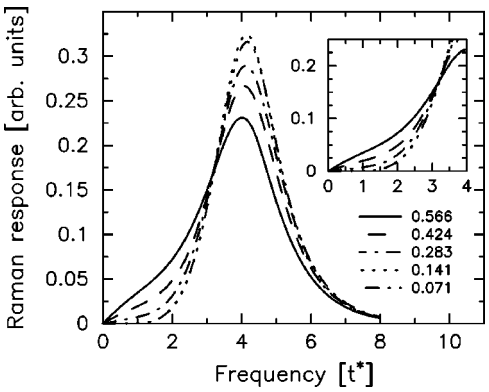

FIG. 2. Nonresonant B_{1g} Raman scattering for a correlated insulator $U=4.24$ and half filling for a number of temperatures ranging from 0.57 to 0.071. Inset is an enlargement of the low-energy features.

where $f(\omega)=1/[1+\exp(\omega/T)]$ is the Fermi function, $A(\epsilon,\omega)=\mathrm{Im}[-1/\pi\{\omega+\mu-\Sigma(\omega)-\epsilon\}]$ is the spectral function, $\rho(\epsilon)$ is the noninteracting density of states on the hypercubic lattice (a Gaussian), and c is a constant.

We study the evolution of the Raman response at half filling, since one can tune the system to move right through the quantum-critical point of the metal-insulator transition. As with other work on the infinite-dimensional Mott transition, we ignore any possible magnetically ordered phases, to examine the behavior near a ''pure'' paramagnetic Mott transition. We analyze the finite-temperature numerical renormalization group calculations.[17] We study three cases here: (i) a correlated insulator just above the transition $U=4.24$ (where the response is ''universal''); (ii) a metal just below the phase transition $U=3.54$ (which undergoes a temperature-dependent metal-insulator transition at $T\approx 0.011$); and (iii) a correlated metal $U=2.12$.

We show the correlated insulator regime in Fig. 2, where we see the expected model-independent behavior of the Raman response: low-energy spectral weight develops at the expense of the higher-energy charge-transfer peak as T increases and a single isosbestic point develops at $\nu\approx U/1.5$. All of the qualitative features of the ''universal behavior'' are shared in the Hubbard model and Falicov-Kimball model solutions. The difference is in the quantitative details, the most apparent one being that at high temperatures in the Hubbard model, the low-energy spectral response does not have a broad peak structure, but rather shows monotonic rising behavior. Note that the model independence arises from (i) the fact that at low temperatures the single-particle density of states (DOS) has the same general shape for any correlated insulator and (ii) the Raman response is just a complicated integral that depends on the single-particle DOS.[9,10] The behavior in the Hubbard model is more characteristic of experiments[2] in FeSi, which has a relatively flat low-energy spectral response. Note that we do not argue that all correlated systems are described by the half-filled Hubbard model—the results shown here verify the model independence of the Raman response on the insulating side of the metal-insulator phase transition, which is observed experimentally in the data of Fig. 1.

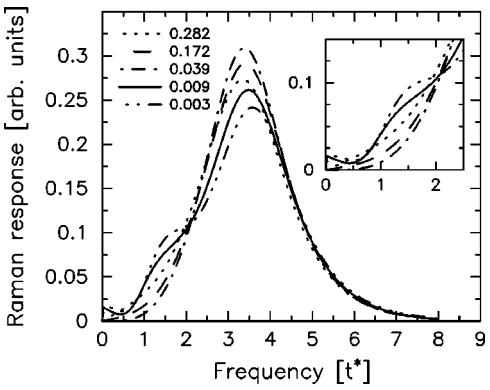

FIG. 3. Nonresonant B_{1g} Raman scattering for a system undergoing a temperature-driven metal-insulator transition $U=3.54$ and half filling for a number of temperatures ranging from 0.28 to 0.003. The inset is an enlargement of the low-energy features.

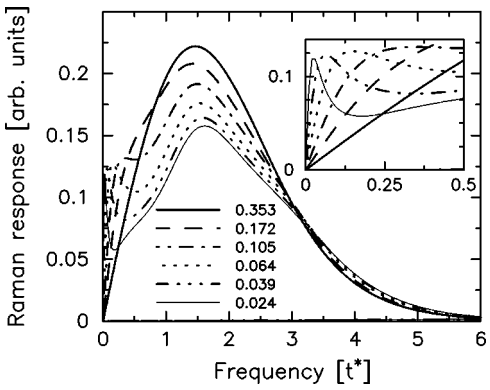

FIG. 4. Nonresonant B_{1g} Raman scattering for a correlated metal $U=2.12$ at half filling for a number of temperatures ranging from 0.35 to 0.024. The inset is an enlargement of the low-energy features.

In Fig. 3, we show results for a system tuned to lie just on the metallic side of the metal-insulator transition (so it undergoes a temperature-driven transition at $T\approx 0.011$). Initially the Raman response acts like an insulator: the charge-transfer peak sharpens and grows in strength, while the low-energy weight is reduced and an isosbestic point appears near $\nu=U/1.7$ as T is lowered. As the temperature is reduced further, the response becomes quite anomalous. Spectral weight shifts back out of the charge-transfer peak into the low-energy region, developing two low-energy bumps and an isosbestic point near $\nu=2$. There is only one experimentally measured system that we know of that shares some of these qualitative features:[1] SmB_6. At low temperatures SmB_6 acts like an insulator, just as seen in Fig. 3, but then at the lowest temperatures ($T<30$ K), a sharp nondispersive peak appears at about 130 cm^{-1}. The qualitative shape of the Raman response is different though, because the weight only grows in a narrow peak, as opposed to the wide frequency range of Fig. 3. We believe it would be interesting to investigate the Raman response of a correlated system that undergoes a similar insulator-metal transition as a function of temperature such as 1% chromium doped vanadium oxide.[18] Such experiments would be feasible if the charge-transfer peak could be pushed to a low enough energy that it lies within the window observable by Raman measurements (which seems likely from the optical conductivity data[18] on insulating vanadium oxide alloys doped close to the metal-insulator transition).

Finally, in Fig. 4, we show the Raman response for a correlated metal at half filling. At high temperatures, the Raman response has a charge-transfer peak centered at $\nu\approx U$. As the temperature is lowered, we see the development and evolution of a low-energy Fermi-liquid peak, which sharpens as T is lowered. This is the classic behavior expected for a correlated metal—at high temperatures there is a large charge-transfer peak centered at an energy just somewhat higher than U, which loses spectral weight as the temperature is lowered and a low-temperature ''metallic'' peak develops at low energy. The Fermi peak has the expected form proportional to $\nu T^2/[T_0(\nu^2+T^4/T_0^2)]$ for a Fermi liquid. The width of the peak (determined by T^2/T_0) decreases as the temperature is lowered and will ultimately vanish at $T=0$. The weight in the metallic peak is much smaller than the weight in the charge-transfer peak. Note that the charge-transfer peak *loses* weight (especially on the low-frequency side) as T is lowered, but there is no low-energy isosbestic point here (in fact an isosbestic point may be developing at $\nu\approx 3.3$). This lack of low-energy isosbestic behavior is unexpected. The development of a large-weight Fermi coherence peak in the interacting density of states seems to destroy the isosbestic behavior.

Unfortunately, we are not aware of any experimental data on a correlated metal that displays this low-temperature development and evolution of the Fermi-liquid peak. Our results show that the consequence of well-defined quasiparticles is the presence of a low-energy peak that grows in intensity and narrows as the temperature decreases. Such a peak might also be observable in materials such as[19,20] $CeSi_2$, $CeSb_2$, $CeBe_{13}$, or $YbAl_3$, which all display the development of a low-temperature Fermi coherence peak at energies less than 100 meV, but may be below the sensitivity of most experiments near the laser line.

To summarize, we have investigated the qualitative features of Raman scattering near the quantum-critical point of a metal-insulator transition (MIT). We confirmed that in the insulating phase, qualitative features are model independent, including an isosbestic point and spectral weight transfer from low to high energy as temperature is lowered. This agrees with all known measurements on correlated insulators including mixed-valence materials, Kondo insulators, and doped antiferromagnets. On the metallic (Fermi-liquid) side of the transition, we found a number of even stranger features that we predict can be seen in systems with a temperature-driven insulator-metal transition (such as the doped vanadium-oxide system). In particular, we saw, that as temperature was lowered, the response first looks like that of an insulator with an isosbestic point and the characteristic spectral-weight shifts. But as T is lowered through the insulator-to-metal transition, there is an abrupt increase in low-energy spectral weight, due to the formation of the Fermi liquid. The low-energy features are complex and increase in size as T is lowered. As one is pushed farther away

from the quantum-critical point to a correlated Fermi liquid, we find that all of the anomalous features disappear. There is no isosbestic point, and a low-energy peak sharpens and moves to lower frequency as T is lowered, characteristic of a Fermi liquid. We propose that such behavior might be observable in a number of cerium-based or ytterbium-based heavy-fermion compounds. We hope our theoretical analysis encourages further experimental work to see these new anomalies.

It is important to note that the nature of a MIT in physical dimensions and its effect on the Raman response are still an open issue: the MIT may be driven by interactions or disorder, may be accompanied by spin or charge ordering, and the DOS at the Fermi level may not be critical, or may vanish smoothly or abruptly at the MIT. In addition, an exact theory for Raman scattering in lower dimensions in correlated electron systems does not yet exist due to computational difficulties. However, since the Raman response on the insulating side of the MIT is governed largely by the presence of the gap in the DOS, different insulating phases might be expected to show the same qualitative features, irrespective of whether or not the MIT is accompanied by charge or spin ordering. This remains a topic of significant interest.

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