Model for a strongly correlated insulator: FeSi

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Prompted by suggestions that FeSi may be a strongly correlated insulator, somewhat analogous to the Kondo insulators, we study the properties of a two-band Hubbard model Hamiltonian in the infinite-dimensional limit. By a calculation of the one-particle self-energy using self-consistent second-order perturbation theory we are able to calculate the temperature dependence of the magnetic susceptibility and of angle-resolved photoemission spectra. The results are in fair agreement with experimental data on FeSi.

I. INTRODUCTION

With the renewed interest in strongly correlated insulators, FeSi has been the subject of renewed experimental and theoretical examination.[1–5] Infrared reflectivity[1] and angle-resolved photoemission[6] measurements are providing a more detailed view of the effects of strong correlations on the electronic excitations in FeSi. In light of these experiments, we have constructed a simplified model for a strongly correlated insulator where the correlation effects can be easily calculated.

In early research, FeSi was identified as a narrow (≈ 50 meV) gap semiconductor with unusual magnetic properties. Its magnetic susceptibility was recognized as inexplicable from simple band-structure models,[7] which would require extremely narrow bands. However, there were no signs of the magnetic ordering which one might expect from such localized states at low temperatures. Since then, explanations for the magnetic behavior in terms of spin fluctuations[8,9] have had reasonable success and have even predicted the existence of a temperature induced magnetic moment, which was found experimentally only much later.[10]

The discovery of the high-temperature superconductors stimulated new interest in the subject of strongly correlated insulators such as CeNiSn, $Ce_3Bi_4Pt_3$, and SmB_6 and FeSi. In the case of FeSi, electronic-structure calculations based on the local-density approximation[2,3] (LDA) find an insulating ground state and elastic properties in reasonable agreement with experiment; however they have concluded that one-electron models are inadequate to explain infrared reflectivity data. Nevertheless, these results have left open the possibility that the true ground state is continuously connected to the one-electron state as the electron-electron interactions measured by a Hubbard U are turned on.[11]

In this paper, we construct a simple model Hamiltonian which appears to contain the physical ingredients needed for understanding the data. We examine our Hamiltonian, a two-band Hubbard model, in the limit of large dimensions. This limit enables us to approximately calculate properties which can be compared to angle-resolved photoemission experiments and magnetic susceptibility measurements. In spite of the fact that the large D limit is effectively a local one, our model predicts strong momentum dependence of the quasiparticle lifetime which should be observable in photoemission experiments, as well as the usual band renormalization effects one expects to find as a result of the correlations. Despite the simplified nature of the model we are able to account for the observed temperature dependence of the magnetic susceptibility and of the optical reflectivity at a semiquantitative level.

II. A MODEL HAMILTONIAN

A. Rationale

Several experimental results guided the structure of our model. The model density of states constructed by Jaccarino *et al.*[7] to fit their susceptibility and specific heat data required a narrow bandwidth of order 50 meV or less for a reasonable fit which is unphysically small without renormalization effects. Schlesinger *et al.* observed a strong temperature dependence of the gap in the optical measurements, which could be ascribed to a loss of coherence. Such effects occur at half-filling in mean field solutions of the Anderson lattice Hamiltonian, a focus of studies on cerium based strongly correlated insulators (e.g., Ref. 11 and references therein). However, we do not expect to find the extremely narrow bands characteristic of f states in a transition metal compound such as FeSi, and indeed, the only anomaly in the specific heat of FeSi matches well with FeSi's magnetic susceptibility and lacks the heavy fermion behavior associated with the Anderson lattice.

LDA calculations for FeSi show a nearly direct insulating gap, surrounded by several narrow iron d bands with rather small contributions from the silicon orbitals.

Thus we were led to construct the following two-band Hamiltonian:

$$H = \sum_{\langle ij\rangle\sigma} -t(\hat{c}^{\dagger}_{i1\sigma}\hat{c}_{j1\sigma} - \hat{c}^{\dagger}_{i2\sigma}\hat{c}_{j2\sigma}) + \sum_{i\sigma} v(\hat{c}^{\dagger}_{i1\sigma}\hat{c}_{i2\sigma} + \text{H.c.}) + \sum_{i} U(\hat{n}_{c_{i1\uparrow}}\hat{n}_{c_{i1\downarrow}} + \hat{n}_{c_{i2\uparrow}}\hat{n}_{c_{i2\downarrow}}). \quad (1)$$

Here we have coupled two Hubbard bands $\hat{c}_{ij\sigma}$, which may be thought of as the iron d bands, with a momentum independent hybridization v. Choosing hopping terms of opposite sign provides a simple direct gap.

In contrast to the Anderson lattice Hamiltonian's conduction band and strongly correlated f band, our model has two symmetric tight binding bands, with a Hubbard U term acting within each band independently. Note that a model with an additional on-site repulsion $U'/2(\hat{n}_{c_{i1}} + \hat{n}_{c_{i2}})(\hat{n}_{c_{i1}} + \hat{n}_{c_{i2}} - 1)$ would shift the energy level at half-filling to a middle Hubbard band, leaving the intraband U to dominate the physics. For simplicity we do not include U' in the model. Instead, our form of intraband U focuses on the competition between U and v. For $U = 0$ the two bands mix and create a direct gap of magnitude $2v$. The ground state simply consists of doubly occupying the states in the lower hybridized band. As $U \to \infty$ the electrons half fill each orbital to avoid double occupancy.

B. Further approximations: $D = \infty$ and self-consistent perturbation theory

The large D approximation, where D is the number of dimensions, introduced by Metzner and Vollhardt[12] to study the Hubbard model, naturally supports several common approximations (e.g., the "local" approximation, Gutzwiller's approximation, and some slave boson results). These approximations underlie our understanding of the heavy fermions, and so we chose the large D limit for study. Using self-consistent second-order perturbation theory for small and intermediate U, Schweitzer and Czycholl[13] found only minor corrections in $1/D$ for the Anderson lattice Hamiltonian for $D = 2$, and found that the corrections compared well with exact results for $D = 1$. We chose to follow their approach, as FeSi's structure is three dimensional, and the renormalization effects we seek are moderate. Note that in this approach no allowance is made for possible symmetry breaking (e.g., charge or spin density waves) in the ground state.

The first major simplification comes in the unperturbed density of states. The energies of the two hybridized tight binding bands are $\tilde{\epsilon}(\mathbf{k}) = \pm\sqrt{v^2 + \epsilon(\mathbf{k})^2}$, where $\epsilon(\mathbf{k}) = -t\sum_{i=1}^{D}\cos(k_i)$. To keep the bandwidth of the unhybridized states the same, we should set $t = t^*/\sqrt{2D}$. This is easily seen[14] by considering the density of states for a single tight binding band,

$$\rho_{TB}(\varepsilon) = \prod_{i=1}^{D}\int_{-\pi}^{\pi}\frac{dk_i}{2\pi}\,\delta\left(\varepsilon - \sum_{j}^{D} -2t\cos k_j\right) = \int\frac{d\tau}{2\pi}e^{i\varepsilon\tau}\prod_{i=1}^{D}\int_{-\pi}^{\pi}\frac{dk_i}{2\pi}e^{-2it\tau\cos k_i} = \int\frac{d\tau}{2\pi}e^{i\varepsilon\tau}\exp[-2D\tau^2t^2 + O(t^4)]. \quad (2)$$

By performing a cumulant expansion in τ, the density of states is reduced to a Gaussian to leading order in D for a single band. We measure all energies in units of t^*, and then for $U = 0$, we exactly diagonalize the Hamiltonian. The resulting density of states is singular, with a gap of $\Delta_0 = 2v$ (in contrast to the indirect gaps $\delta \propto {v_R}^2/D$ in the Anderson lattice Hamiltonian where v^R is a renormalized hybridization and D the bandwidth),

$$\rho_0(\varepsilon) = \frac{1}{\sqrt{\pi}}\frac{\varepsilon}{\sqrt{\varepsilon^2 - v^2}}\exp(-\sqrt{\varepsilon^2 - v^2}) \quad (|\varepsilon| > v). \quad (3)$$

For $U \neq 0$ we incorporate the effects of interactions on the density of states, as well as other properties, by calculating the self-energy $\mathbf{\Sigma}(\mathbf{k},\omega)$ of the particles. This determines the full Green's function via Dyson's equation,

$$\mathbf{G}(\mathbf{k},\omega) = [\mathbf{G}_0^{-1}(\mathbf{k},\omega) - \mathbf{\Sigma}(\mathbf{k},\omega)]^{-1}. \quad (4)$$

Searching for a self-consistent solution for this in perturbation theory, we simplify the form of $\mathbf{\Sigma}(\mathbf{k},\omega)$ by noting we can choose $\Sigma_{11} = \Sigma_{22}$ and $\Sigma_{12} = \Sigma_{21}$, because Σ_{ij} will, to second order, depend only on G_{ij}, and $\epsilon_1(\mathbf{k}) = -\epsilon_2(\mathbf{k})$.

The first order terms in the expansion correspond to Hartree-Fock theory; they simply represent the effect of a mean background field upon the particles. If we assume a paramagnetic state, then at half-filling $\Sigma^{(1)}_{ii,\sigma}(\mathbf{k},\omega) = Un_{i\bar{\sigma}} = U/2$.

For the second order terms we take advantage of the large D limit, in which the self-energy becomes momentum independent. A standard diagrammatic expansion yields

$$\Sigma^{(2)}_{ij,\sigma}(\mathbf{k},i\omega_n) = -U^2\beta^2\sum_{\substack{i\omega_1,i\omega_2,\mathbf{k}_1\\ \mathbf{k}_2,\mathbf{k}_3}} G_{ij,\bar{\sigma}}(\mathbf{k}_2,i\omega_1)G_{ij,\bar{\sigma}}(\mathbf{k}_3,i\omega_2)G_{ij,\sigma}(\mathbf{k}_1,i\omega_n - i\omega_1 + i\omega_2)\delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k})$$
$$= -U^2\beta^2\sum_{i\omega_1,i\omega_2}\int d\epsilon_1 d\epsilon_2 d\epsilon_3 G_{ij,\bar{\sigma}}(\epsilon_2,i\omega_1)G_{ij,\bar{\sigma}}(\epsilon_3,i\omega_2)G_{ij,\sigma}(\epsilon_1,i\omega_n - i\omega_1 + i\omega_2)w(\mathbf{k};\epsilon_1,\epsilon_2,\epsilon_3), \quad (5)$$

$$w(\mathbf{k};\epsilon_1,\epsilon_2,\epsilon_3) = \sum_{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}\delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k})\prod_{i=1}^{3}\delta(\epsilon_i - \epsilon(\mathbf{k}_i)).$$

If we express the delta functions in $w(\mathbf{k};\epsilon_1,\epsilon_2,\epsilon_3)$ as integrals and sums over exponentials as before, we find that to leading order in $1/D$, $w(\mathbf{k};\epsilon_1,\epsilon_2,\epsilon_3)=1$, and thus Σ_{ij} is momentum independent. This result is true to all orders in perturbation theory.[14]

In evaluating the second order corrections to $\Sigma_{11}(\omega)$ and $\Sigma_{12}(\omega)$ (again, assuming a paramagnetic state), the Lehmann spectral representation of the Green's function,

$$G_{ij}(\mathbf{k},z)=-\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\omega\,\mathrm{Im}\,G_{ij}(\mathbf{k},\omega+i\epsilon)}{z-\omega}, \tag{6}$$

defines the Green's function over the complex plane in terms of its values for real frequencies. Our expression for $\Sigma^{(2)}(\omega)$ only depends on the on-site real space component, so we define,

$$\begin{aligned}\bar{G}_{ij}(\omega+i\epsilon)&=\sum_{\mathbf{k}}\mathrm{Im}\,G_{ij}(\mathbf{k},\omega+i\epsilon)\\&=\frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}d\epsilon_{\mathbf{k}}\exp(-\epsilon_{\mathbf{k}}^2)\,\mathrm{Im}\,G_{ij}(\epsilon_{\mathbf{k}},\omega+i\epsilon).\end{aligned} \tag{7}$$

$\bar{G}_{ij}(\omega+i\epsilon)$ can be evaluated in terms of Fadeeva's function (see the Appendix), which has generally available numerical implementations.[15] Then we evaluate the sums over Matsubara frequencies,

$$\Sigma_{ij}^{(2)}(i\omega_n)=-\frac{U^2}{\pi^3}\int dz_1dz_2dz_3\bar{G}_{ij}(z_1)\bar{G}_{ij}(z_2)\bar{G}_{ij}(z_3)\frac{f(z_1)f(z_2)[1-f(z_3)]+[1-f(z_1)][1-f(z_2)]f(z_3)}{z_1+z_2-z_3-i\omega_n}. \tag{8}$$

If we analytically continue the self-energy ($i\omega_n\to\omega+i\epsilon$) then the energy denominator can be rewritten as an integral over an exponential. The resulting expression couples functions of the Green's functions only through a Fourier transform which we evaluate numerically:

$$\begin{aligned}\Sigma_{ij}^{(2)}(\omega)=-\frac{iU^2}{\pi^3}\int_0^{\infty}d\tau e^{i\omega\tau}[&\alpha_{ij}(\tau)\alpha_{ij}(\tau)\beta_{ij}(-\tau)\\&+\beta_{ij}(\tau)\beta_{ij}(\tau)\alpha_{ij}(-\tau)].\end{aligned} \tag{9}$$

Here $\alpha_{ij}(\tau)$ and $\beta_{ij}(\tau)$ are Fourier transforms of the site diagonal Green's functions,

$$\begin{aligned}\alpha_{ij}(\tau)&=\frac{-1}{\pi}\int_{-\infty}^{\infty}d\varepsilon\;e^{-i\tau\varepsilon}\bar{G}_{ij}(\varepsilon+i\epsilon)f(\varepsilon),\\\beta_{ij}(\tau)&=\frac{-1}{\pi}\int_{-\infty}^{\infty}d\varepsilon\;e^{-i\tau\varepsilon}\bar{G}_{ij}(\varepsilon+i\epsilon)[1-f(\varepsilon)].\end{aligned} \tag{10}$$

To solve these equations self-consistently, we start with a null self-energy, and calculate the corrections according to Eq. (9). The succeeding values of the self-energy are linear combinations of the current value and the new calculation. By gradually mixing in the new solution, we avoid oscillatory behavior. When mixing in 20% of the new solution with the old, we find convergence within 10 iterations. Our choice of mesh size for the discrete Fourier transform produced results consistent with larger mesh sizes. The evaluation of the Green's function at frequency $\omega+i\epsilon$ in Eq. (4) is performed numerically; by keeping $i\epsilon$ finite we force a finite lifetime for all states.

With a self-consistent $\Sigma_{ij}(\omega)$ we calculate several physical quantities. The spectral function $A(\epsilon_{\mathbf{k}},\omega)=-\frac{1}{\pi}\mathrm{Im}\,G(\epsilon_{\mathbf{k}},\omega)$ and the one-particle density of states $\rho(\omega)=\sum_{\mathbf{k}}A(\epsilon_{\mathbf{k}},\omega)$ can be compared to angle-resolved and angle-integrated photoemission studies. The static and dynamic susceptibilities are also easily calculated in the large D limit. For all momenta except for $\mathbf{q}=\mathbf{0}$ and $\mathbf{q}=\mathbf{Q}$, if we exclude vertex corrections, the momenta decouple and the two-particle response function is simply the product of the one-particle functions. In the case of the optical conductivity, this decoupling, along with the parity of the current operator were shown by Khurana[16] to force the vertex corrections to vanish. Thus to compare our model with optical conductivity measurements, we need only compute the joint density of states:

$$\sigma(\omega,T)=\int d\epsilon\rho(\epsilon)\rho(\epsilon+\omega)[1-f(\epsilon+\omega,T)]. \tag{11}$$

While vertex corrections are nonzero for the magnetic susceptibility,[17] for our system we expect no magnetic instabilities, and so approximate the magnetic susceptibility $\chi(T)$ for small to to intermediate U as simply

$$\chi(T)\propto\lim_{\omega\to0}\sigma(\omega,T)/T. \tag{12}$$

III. RESULTS

Our model has two adjustable parameters, v and U measured in units of the hopping parameter t^*. To compare our model with physical properties in FeSi, we use our LDA calculation of the band gap (≈ 150 meV) relative to the width of the d bands (≈ 800 meV) to choose $v=0.125$. Our choice of suitable values for U is based upon consistency between our results and experimental observations.

Figure 1 depicts a calculation of the self-energy for a range of temperatures. Our calculation satisfies Luttinger's theorem, as $\mathrm{Im}\,\Sigma_{11}(\omega)=0$ at the Fermi energy at zero temperature. While calculating the corrections to the self-energy is the heart of our calculation, it is dif-

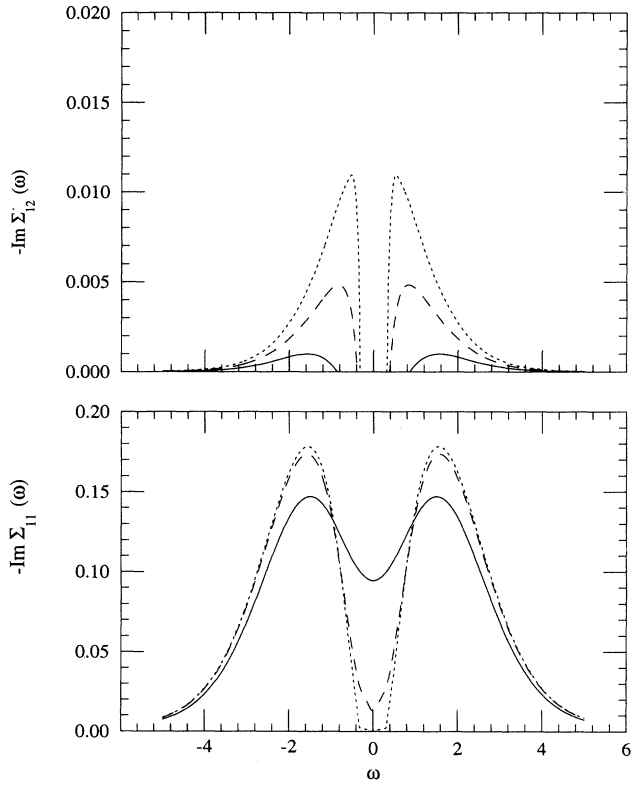

FIG. 1. Diagonal (bottom) and off-diagonal (top) components of the imaginary part of the self-energy $\Sigma_{ij}(\omega)$ as a function of temperature. Solid, dashed, and dotted lines correspond to $T = 0.33$, 0.10, and 0.033, respectively.

ficult to compare to experimental results, so we consider quantities such as the density of states (DOS).

From the DOS we see the gap is preserved, but reduced by the interactions at zero temperature. Because the boundary of the gap is singular for $U = 0$ [Eq. (3)], we define the gap size as the separation between the two maximal values in the DOS. We find a fair empirical fit for the gap Δ, as a function of v and U,

$$\Delta(v, U) = \Delta_0 v^r / v, \tag{13}$$

where

$$v^r = \frac{v}{\sqrt{1 + 0.4U^2/v}} \tag{14}$$

is a renormalized coupling (Fig. 2). This confirms our expectations of a competition between U^2 (because corrections are second order in U) and v to reduce the gap.

If we look at the spectral function for a fixed value of $\epsilon_{\mathbf{k}}$ close to the band edge, we find a familiar scenario (Fig. 3). Our model exhibits a quasiparticle pole, with an incoherent background of width $\approx U$ forming above a threshold at v. By numerically fitting the quasiparticle pole to a Lorentzian, $\alpha\Gamma/[(\omega-\omega_0)^2+\Gamma^2]$, we can study the dependence of the quasiparticle's energy ω_0, scattering rate Γ, and weight α on ϵ_k and T.

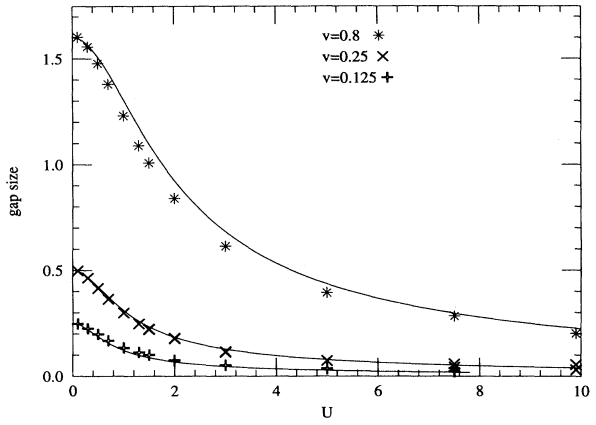

FIG. 2. Size of the gap Δ, in the one-particle spectral function $-\frac{1}{\pi}\,\mathrm{Im}\,G(\omega)$ as a function of U for selected values of v. Δ is the distance between maximal values of the spectral function. Solid lines show the empirical formula in Eq. (13).

Examining the dispersion of the quasiparticle energy, we find it is strongly renormalized by U, and that the overall bandwidth is reduced by roughly v^r/v (Fig. 4). For moderate temperatures, the temperature dependence of this dispersion is relatively minor (Fig. 5).

The scattering rate of the quasiparticles near the Fermi energy exhibits a simple temperature dependence, with $\Gamma \propto \exp(-\beta\Delta)$. Thus the lifetime of a quasiparticle is determined by scattering off of thermally excited particles and holes near the Fermi energy. However, this scattering rate is strongly dependent on ϵ_k. As the quasiparticle energies reach the energy scale of particle-hole pair creation, the lifetime dramatically shortens (Fig. 6).

The distribution of spectral weight between the quasiparticle peak and the incoherent background is strongly temperature dependent (Fig. 3). At low temperatures,

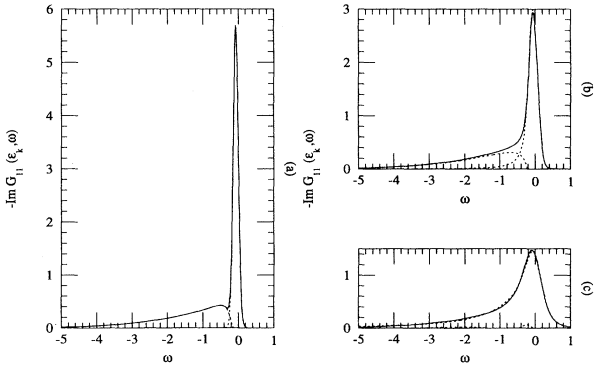

FIG. 3. One-particle spectral function for $\epsilon_{\mathbf{k}} = 0$, $U = 1, v = 0.125$ at selected temperatures: $T = 0.067\Delta_0$ (a), $T = 0.4\Delta_0$ (b), and $T = 1.3\Delta_0$ (c). Dotted lines indicate a least squares fit of the quasiparticle peak to a Lorentzian broadened by a Fermi function and residuals from incoherent scattering. Line shapes have been convolved with a Gaussian to reflect instrumental broadening.

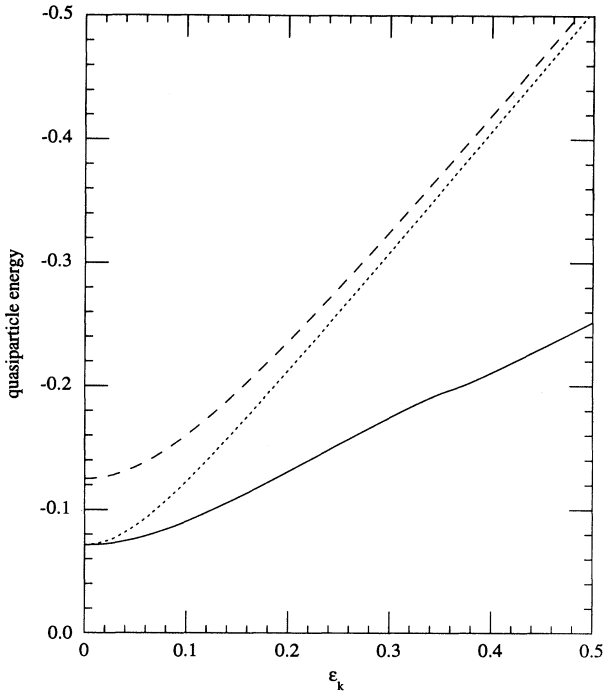

FIG. 4. Dispersion of the fitted quasiparticle peak for $U = 1$ (solid) and $U = 0$ (dashed). Dotted line illustrates the dispersion which would occur if the only effect of U were to renormalize v.

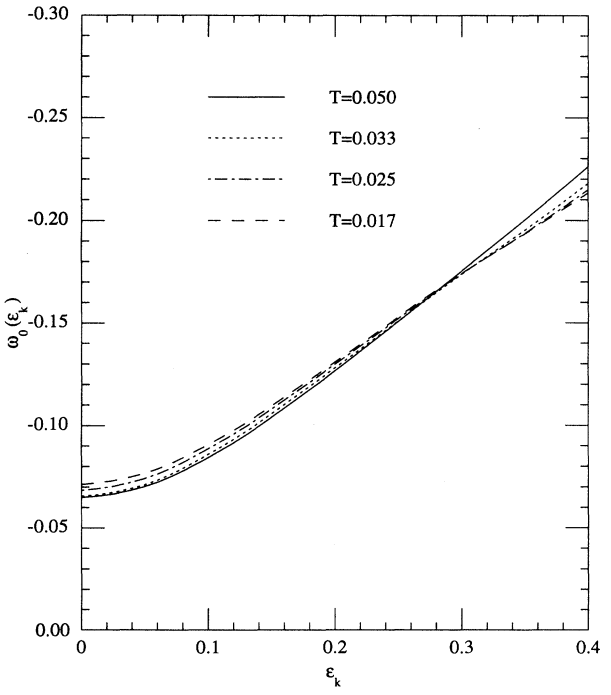

FIG. 5. The k dependence of the fitted quasiparticle energy for $U = 1$ for various temperatures. Above $T = 0.05$ the peak in the spectral function fits poorly to a Lorentzian, and is unsuitable for comparison.

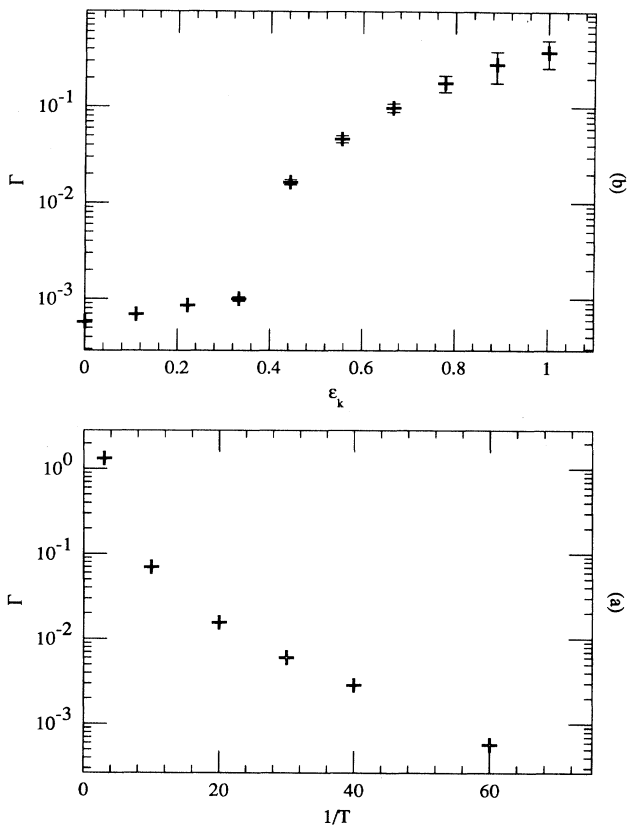

FIG. 6. Inverse lifetime of the quasiparticle for $U = 1, v = 0.125$, as a function of T at $\epsilon_k = 0$ (a) and ϵ_k (b).

the weight is divided between the two and remains roughly constant. However, as the temperature increases, the spectral weight rapidly shifts to the broadened quasiparticle pole until that is all that remains visible. While there is some uncertainty in the magnitude of this effect as a result of our fitting procedures, it is strong enough that the shift in spectral weight is clearly established.

The two-particle Green's functions provide other tests of our model. The static susceptibility, which we approximate by $\chi(T) = \sigma(0,T)/T$ provides a nice illustration of the effects of the on-site correlation. In Fig. 7 we have plotted $\chi(T)$ for the interacting and noninteracting cases, respectively, and compared them to experimental data.[10] By trying different values of U, we find the best semiquantitative agreement for $\chi(T)$ at $U = 0.36$. Both cases exhibit a gap which eventually turns over to a Curie form $\chi \propto 1/T$, but the finite U case saturates more quickly and provides a much better fit to the data.

In Fig. 7 by scaling $\chi(T)$ we connect the energy scales of our theory to experiment. For this scale, the natural energy unit (which comes from the hopping term t) corresponds to an energy of 0.7 eV. This leads to a renormalized gap size of about 70 meV, and a noninteracting one of about 140 meV for the above choices of v and U. These correspond well to the experimental findings and to the LDA calculations of FeSi, respectively.

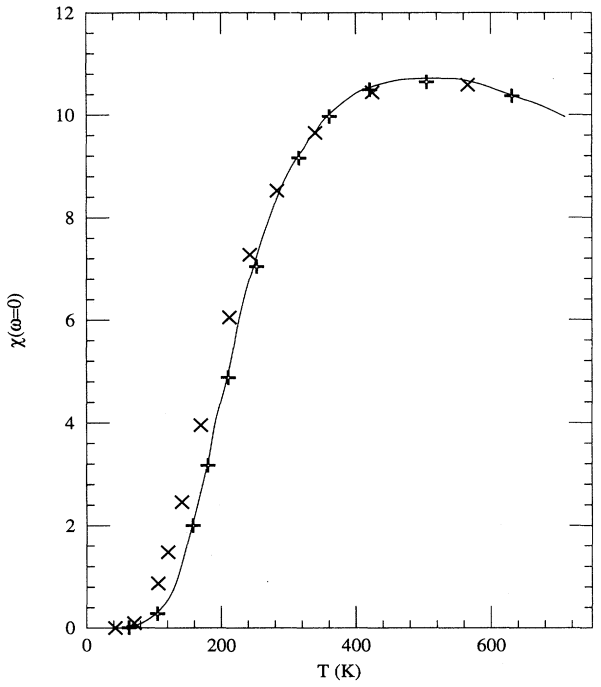

FIG. 7. Magnetic susceptibility $\chi(T)$ of our model for $U = 0.0$ (crosses) and $U = 0.36$ (plus signs) as a function of temperature. The solid line indicates experimental measurements on FeSi from Jaccarino *et al.* (Ref. 7).

Another experimental test is the optical conductivity. In Fig. 8 we show the joint density of states as a function of temperature. For this measurement we have chosen $U = 1$ as it seems to give better qualitative agreement with the trends observed in the photoemission data of Park *et al.*[6] At low temperatures we have a gap indicating the separation between the two quasiparticle bands and a secondary band created by transitions to states in the background. As we increase the temperature, the conductivity smears out, and the spectral weight becomes

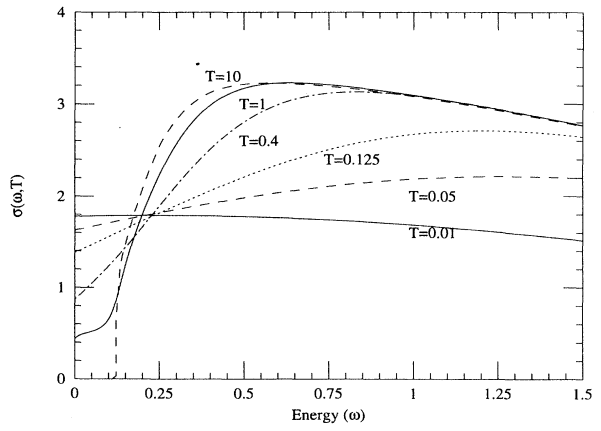

FIG. 8. Joint density of states $\sigma(\omega, T)$. Different curves indicate temperature, for $v = 0.125$ and $U = 1.0$.

distributed over a much larger range in energies. While there is some shift of spectral weight to within the gap, the reduction of the main peak far exceeds this as it is distributed across all temperatures. The gap fills in uniformly, with significant filling by $T \approx \Delta/2$, in contrast to models where there are free carriers in a Drude peak.[3]

IV. DISCUSSION

In this paper we have begun exploring the possibilities of a simple model Hamiltonian for modeling the narrow gap semiconductor FeSi, using the large D limit to compare with specific experimental results. The large D limit provides a simple method of incorporating the effect of electron correlations. After choosing model parameters for the base Hamiltonian consistent with our earlier electronic structure calculations in the LDA, we calculated one- and two-particle properties in self-consistent second order perturbation theory which we believe will provide a reasonable approximation at small to intermediate values of U.

The many-body effects modify the Bloch states resulting in a quasiparticle spectrum with an incoherent background. The effects of the correlations on the quasiparticle peaks lead to a reduction of the gap, which, for moderate values of U, may explain the discrepancies seen between band structure calculations and observed properties of FeSi.

The renormalization of the quasiparticle peaks also manifests itself as an apparent narrowing of the bands themselves. The resulting paramagnetic susceptibility explains the observations of $\chi(T)$ which had long ago been fit to an extremely narrow two-band model. While one might question the need to consider correlation effects given the relatively good fit for the $U = 0$ case in Fig. 7, one must remember that even in the noninteracting state our infinite dimensional model has very narrow bands in the DOS [Eq. (3)]. Nevertheless, the correlation reduced bandwidths are more rapidly saturated as the temperature increases, resulting in a more favorable comparison with experiment.

Several features from our calculation should be very apparent in angle-resolved photoemission experiments (e.g., Ref. 6). The lifetime and spectral weights of the narrow quasiparticle peaks are strongly temperature dependent as a result of particle-hole pair creation. The incoherent portion of the spectral function is also temperature dependent. This could be potentially significant in analyzing photoemission spectra, as it may bias attempts to normalize spectral weights at different temperatures.

However, our model does not reproduce the shifts of spectral weight seen by Schlesinger *et al.* in the optical conductivity. Thus either the $D \to \infty$ limit is inadequate to account for the shifts or there is some question about these experimental findings. Note that because of neglect of vertex corrections, we do not expect the same values of the parameters to work for both $\chi(T)$ and the optical or photoemission measurements. In fact, we find different values of U seem appropriate for $\chi(T)$ and for single-particle properties.

Nonetheless, the simplicity of this model, a strongly correlated, direct gap insulator, should lend itself well to a variety of analytical techniques. Future exploration could be made using Gutzwiller projection operators or calculating finite D corrections. Georges and Kotliar[18] have shown that in the large D limit the Hubbard model can be transformed into an impurity problem with a supplementary self-consistency equation, and which can be solved exactly, even in the limit of large U (where the form of self-consistent second order perturbation theory used here fails). This inspired a great deal of work[19] which should extend to our model as well. All of these offer opportunities to gain further insights in the physics of strongly correlated insulators.

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APPENDIX: LOCAL GREEN'S FUNCTION

For a given frequency, we can evaluate the local Green's function numerically in terms of Fadeeva's function,

$$w(z) = \frac{i}{\pi}\int_{-\infty}^{\infty}\frac{e^{-t^2}dt}{z-t} \quad (\mathrm{Im}\, z > 0), \tag{A1}$$

which has a widely available computer implementation.[15] We start with the Dyson equation, and replace the momentum summation by an integration over the density of states,

$$\begin{aligned} G_0^{-1}(\omega;k) &= \omega\mathbf{I} + \epsilon_k\tau_3 - v\tau_2, \\ \bar{\mathbf{G}}(\omega) &= \mathrm{Im}\sum_{\mathbf{k}}[\mathbf{G}_0^{-1}(\omega;\mathbf{k}) - \mathbf{\Sigma}(\omega)]^{-1}, \\ \bar{\mathbf{G}}(\omega) &= \mathrm{Im}\,\frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty} d\epsilon_k e^{-\epsilon_k^2}[\mathbf{G}_0^{-1}(\omega;\epsilon_k) - \mathbf{\Sigma}(\omega)]^{-1}. \end{aligned} \tag{A2}$$

Because the self-energy is independent of the momenta, the integrals can be expressed in terms of $w(z)$, yielding

$$\begin{aligned} \bar{G}_{11}(\omega) &= \mathrm{Im}\,\frac{2}{\sqrt{\pi}}[z - \Sigma_{11}(\omega)]\zeta(\omega), \\ \bar{G}_{12}(\omega) &= \mathrm{Im}\,\frac{2}{\sqrt{\pi}}[\Sigma_{12}(\omega) - v]\zeta(\omega), \\ \zeta(\omega) &= -i\pi\frac{w\{\sqrt{[\omega - \Sigma_{11}(\omega)]^2 - [v - \Sigma_{12}(\omega)]^2}\}}{[\omega - \Sigma_{11}(\omega)]^2 - [v - \Sigma_{12}(\omega)]^2}, \end{aligned} \tag{A3}$$

where we have taken the root within the domain of Eq. (A1).

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[1] Z. Schlesinger *et al.*, Phys. Rev. Lett. **71**, 1748 (1993).
[2] L. Mattheiss and D. Hamann, Phys. Rev. B **47**, 13 114 (1993).
[3] C. Fu, M. Krijn, and S. Doniach, Phys. Rev. B **49**, 2219 (1994).
[4] J. Sarrao *et al.*, Physica B **199&200**, 478 (1994).
[5] M.A. Continentino, G.M. Japiassu, and A. Troper, Phys. Rev. B **49**, 4432 (1994).
[6] C. Park *et al.* (unpublished).
[7] V. Jaccarino *et al.*, Phys. Rev. **160**, 476 (1967).
[8] Y. Takahashi, M. Tano, and T. Moriya, J. Magn. Magn. Mater. **31-34**, 329 (1983).
[9] S. Evangelou and D. Edwards, J. Phys. C **16**, 2121 (1983).
[10] K. Tajima, Y. Endoh, J. Fisher, and G. Shirane, Phys. Rev. B **38**, 6954 (1988).
[11] S. Doniach, C. Fu, and S. Trugman, Physica B **199&200**, 450 (1994).
[12] W. Metzner and D. Vollhardt, Phys. Rev. Lett. **62**, 324 (1989).
[13] H. Schweitzer and G. Czycholl, Solid State Commun. **74**, 735 (1990).
[14] E. Müller-Hartmann, Z. Phys. B **74**, 507 (1989).
[15] M. Abramowitz and I. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1972); G. Poppe and C. Wijers, ACM Trans. Math. Soft **16**, 38 (1990).
[16] A. Khurana, Phys. Rev. Lett. **64**, 1990 (1990).
[17] V. Zlatić and B. Horvatić, Solid State Commun. **75**, 263 (1990).
[18] A. Georges and G. Kotliar, Phys. Rev. B **45**, 6479 (1992).
[19] A. Georges and W. Krauth, Phys. Rev. B **48**, 7167 (1993); X.Y. Zhang, M. Rozenberg, and G. Kotliar, Phys. Rev. Lett. **70**, 1666 (1993); M. Jarrell, *ibid.* **69**, 168 (1992).