earlier—cf. also Refs. 12 and 12a. (The accurate calculation at $\tilde{k} \to 0$ accounts for a small difference between Γ^* and Γ.) In Eq. (15), $t \sim 1$ for $D=1$, $t \sim |V^0| \ln(a/d_0)$ for $D=2$, and $t \sim |V^0| d_t^{2-D}$ for $D \geqslant 3$. By Eqs. (10)–(12), (15), and (16), the localization lengths are very different for $\langle T \rangle$, $\langle T^{-1} \rangle$, $\langle (TT^+)^{-1} \rangle$, and resistance.[2,4]

If the impurity size is $d_0 \neq 0$ in all directions, then, to allow for the nonoverlapping impurity x projections, d_0 must be less than the average interimpurity distance $\tilde{d}_0 \sim 1/n_i S$. Until $\tilde{d}_0 > a$, i.e., until $n_i Sa < 1$, the finite value of d_0 should not be important. But when $n_i S$ is large enough, $n_i Sa \gg 1$, then the "allowed" impurity size $d_0 < (n_i S)^{-1}$ provides a single-impurity attenuation, which tends to zero when $d_0 \to 0$ (and $D \geqslant 2$). This may signal the crucial role for $D \geqslant 2$ of the assumed nonoverlapping of impurity projections. Another very important problem is how much the results of this section depend on the scattering model (13). The presented approach is straight-forwardly generalized to any (e.g., electromagnetic wave) scattering.

I am very grateful to Professor P. Lee for the skepticism which forces me to find the exact solution; to Professor B. Halperin for the suggestion of periodic boundary conditions; and to Professor P. Anderson and Professor P. Platzman for valuable discussions on a long-range correlation.

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[15] Matrices θ account for the states both with real and with imaginary wave vectors (the latter exponentially decay in the vicinity of the impurity).

[15a] In a periodic (along x) lattice, a band is determined by the eigenvalues $\exp(-i\tilde{k}a)$ of $\omega_j \omega_{j+1}^{-1} \theta^0$; $\tilde{k}$ is a quasi wave vector, and a is a period.

[16] Equation (6) is valid for $U \propto \delta(x)$. If the impurity size is d_0, then L in Eq. (6) should be replaced by $L - Nd_0$ and $x_{j+1} - x_j$ by $x_{j+1} - x_j - d_0$.

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Optical Structure near 20 meV in Valence-Fluctuation Compounds

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The first measurement is reported of low-energy (20 meV) structure in the low-temperature dielectric function of two valence-fluctuation materials: $CePd_3$ and $YbCu_2Si_2$. The structure is consistent with energy-dependent scattering of electrons off a resonant level whose width and position (relative to the Fermi level) are roughly comparable. No such structure is observed in the integral-valent materials YPd_3, $DyPd_3$, and $LuCu_2Si_2$. The valence-fluctuation compounds $CeCu_2Si_2$ and $CeAl_3$ do not show a resonance above 4 meV.

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In this Letter we report far-infrared optical-absorption measurements at photon energies between 4 and 40 meV in the valence-fluctuation compounds $CePd_3$ and $YbCu_2Si_2$. Both compounds show a broad absorption feature at low temperature, which we attribute to electron scattering from a resonant level some 20 meV away from the Fermi level. No such structure exists in the integral-valent analog compounds YPd_3, $DyPd_3$, and $LuCu_2Si_2$.

Polycrystalline samples of RPd_3 (R = Ce, Y, Dy), RCu_2Si_2 (R = Ce, Yb, Lu), and $CeAl_3$ were prepared by arc melting stoichiometric amounts of the constituents in an argon atmosphere. The ingots were annealed as follows: one week at 850 °C for YPd_3 and $DyPd_3$, one week at 800 °C for the RCu_2Si_2 compounds, and four weeks at 900 °C for $CeAl_3$. The $CePd_3$ sample did not require annealing. All of the samples were found to consist of a single phase according to x-ray diffraction analysis. Slices of about 0.5 mm thick were cut from the ingots and mechanically polished or chemically etched to prepare the surface for the optical measurements.

We measure the optical absorptivity A (the fraction of incident radiation absorbed by the metal on a single reflection at normal incidence) using a dual-cavity technique.[1] Radiation is coupled through a small hole into a brass cavity partially lined with the sample, and makes on average about 100 reflections before exiting through a second hole to the detector. An identical brass cavity serves as a reference. We have refined an approximate analysis of microwave cavities by Lamb[2] and applied it to extract absolute quantitative values of the absorptivity from our dual-cavity measurements. The details have been given elsewhere.[3]

High-resistivity metals at low frequencies are in the classical skin-effect regime,[4] where the absorptivity can be expressed in terms of the complex magnetic permeability $\mu(\omega)$ and the complex dielectric function $\epsilon(\omega)$ as[5]

$$A(\omega) = 4\mathrm{Re}[\mu(\omega)/\epsilon(\omega)]^{1/2} . \qquad (1)$$

Usually μ can be taken to be 1. For a free-electron metal modeled by the Drude theory with an energy-independent electronic relaxation time τ, Eq. (1) simplifies in the limit $\omega\tau \ll 1$ to

$$A(\omega) = (8\omega/\omega_p{}^2\tau)^{1/2} , \qquad (2)$$

where ω_p is the plasma frequency. In Fig. 1 the absorptivity of $CePd_3$ as a function of frequency at various temperatures is compared to the predicted Drude response of $CePd_3$ at 4.2 K, calculated from the measured dc electrical resistivity of 41 $\mu\Omega$ cm. Also shown is the low-temperature absorptivity of YPd_3, an isostructural integral-valent analog of $CePd_3$. YPd_3 does indeed show the Drude behavior, rising slowly with a negative curvature as the frequency increases. Similar results are seen for $DyPd_3$. For both YPd_3 and $DyPd_3$ the temperature dependence is weak. The low-temperature absorptivity of $CePd_3$, however, departs radically from the Drude behavior with increasing frequency, rising with a positive curvature to a maximum at 19 meV about 2.5 times greater than the value predicted by the Drude theory. The anomaly is also strongly temperature dependent, decreasing with increasing temperature up to 150 K, where the absorptivity once again appears Drude-like. The sharp secondary feature at 15 meV will be discussed later.

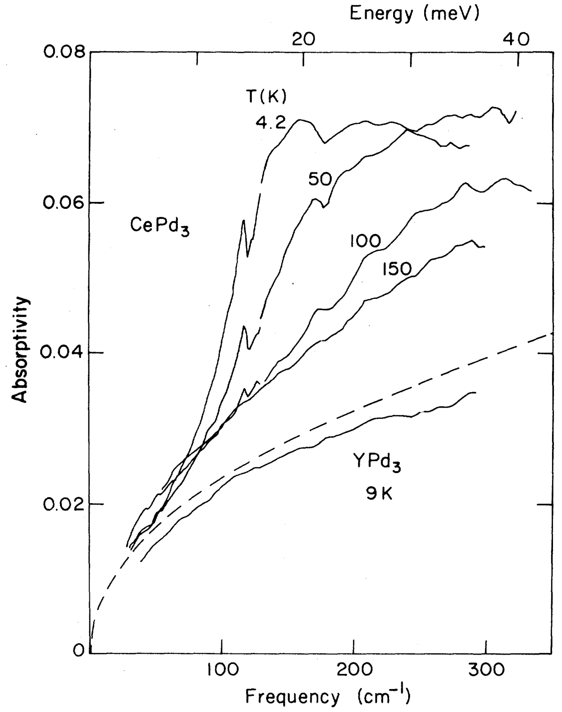

FIG. 1. Absorptivity vs frequency for $CePd_3$ and YPd_3 at various temperatures. The instrumental resolution is 1 meV above 16 meV and 0.4 meV below 16 meV. The dashed curve is the predicted Drude response of $CePd_3$ at 4.2 K.

We can express the dielectric function $\epsilon = \epsilon_1 + i\epsilon_2$ corresponding to $A(\omega)$ in a Drude-like form provided we let both τ and ω_p become frequency dependent quantities $\tau(\omega)$ and $\omega_p(\omega)$[6]:

$$\epsilon_1 = -\omega\tau\epsilon_2 , \qquad (3)$$
$$\epsilon_2 = \omega_p{}^2\tau/[\omega(1+\omega^2\tau^2)] .$$

In the limit $\omega\tau \ll 1$, ϵ_1 is negligible compared to ϵ_2, and ϵ_2 is just $\omega_p{}^2\tau/\omega$. Since we know only one quantity over a frequency region too restricted to perform a reliable Kramers-Kronig analysis, we have no way of separating the product $\omega_p{}^2\tau$, which we therefore write as an effective relaxation time $(\omega_p{}^2\tau)_e$. Figure 2 shows $(\omega_p{}^2\tau)_e{}^{-1}$ for $CePd_3$ at 4.2 K calculated from $A(\omega)$ by inverting Eq. (2). We write the effective relaxation time as

$$(\omega_p{}^2\tau)_e{}^{-1} = (\omega_p{}^2\tau)_0{}^{-1} + (\omega_p{}^2\tau)_f{}^{-1}. \tag{4}$$

The energy-independent term $(\omega_p{}^2\tau)_0$ describes the scattering in normal metals. Its value, indicated in Fig. 2, is taken from the measured dc electrical resistivity of $LaPd_3$.[7] The second contribution $(\omega_p{}^2\tau)_f$ describes the additional energy-dependent scattering in valence-fluctuation materials. Figure 2 suggests an $(\omega_p{}^2\tau)_f$ of the form

$$(\omega_p{}^2\tau)_f{}^{-1} = \frac{S(\Gamma/2)^2}{(\hbar\omega - E_0)^2 + (\Gamma/2)^2}. \tag{5}$$

This is the form of a resonant scattering level at an energy E_0 above the Fermi level with a full width Γ and strength S. The temperature dependence of the absorptivity enhancement is qualitatively consistent with this model, since the anomaly weakens at temperatures where the resonant level is expected to be thermally populated. The origin of the resonant level is not clear. It may be the Ce f^1 level, albeit at a very low energy. Alternatively, it may represent scattering from a phonon mode associated with the valence fluctuation.

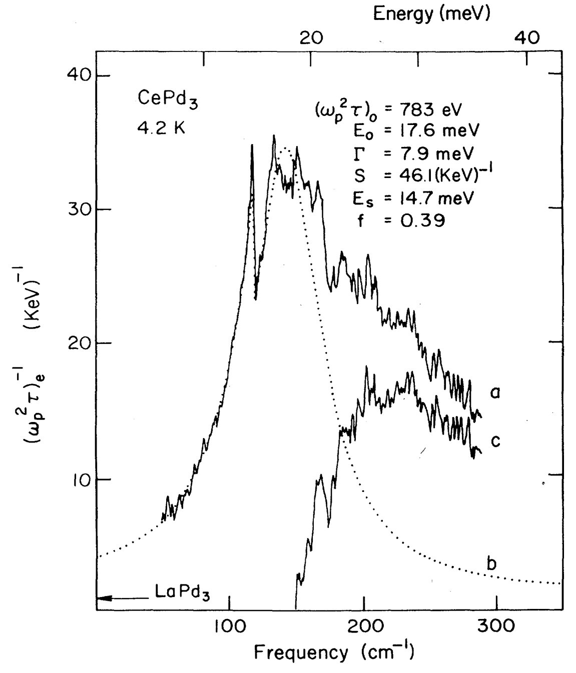

FIG. 2. Curve *a*, inverse effective relaxation time vs frequency for $CePd_3$ at 4.2 K. The resolution is 0.2 meV. Curve *b*, fit to the data with use of the resonant-level model [Eq. (5)] with a step in the conductivity [Eq. (6)]. The parameters are listed in the figure. Curve *c*, difference between the data and the model fit.

The fit of Eq. (5) to $(\omega_p{}^2\tau)_e{}^{-1}$ for $CePd_3$ is complicated by the secondary structure at 15 meV. If we initially restrict our attention to the region of positive curvature below 15 meV, an excellent fit to the broad resonance is obtained with $E_0 = 17.6$ meV and $\Gamma = 7.9$ meV, as shown by the dotted curve in Fig. 2. The strength $S = 46.1$ $(\text{keV})^{-1}$ is consistent with maximal incoherent scattering by each Ce ion. More generally, however, we must take into account the secondary spike at 15 meV. The high-resolution (0.2 meV) data of Fig. 2 shows that the spike is remarkably sharp at 4.2 K, with a full width of about 0.4 meV, and is highly asymmetric, dropping off on the high-frequency side at the resolution of the data. The magnitude decreases with increasing temperature, as seen in Fig. 1, but the position and width do not change appreciably with temperature. At all temperatures above 4.2 K the width is less than kT. There is no detectable shift in the position, width, or magnitude of this feature in magnetic fields up to 75 kG at a resolution of 0.03 meV, implying a nonmagnetic origin for the spike. We have investigated possible model fits to this feature using an optic phonon mode or a sudden increase in the ac optical conductivity. A transverse-optic phonon produces a resonance in $\epsilon(\omega)$ with a strength determined by ω_{PM}, the plasma frequency for ions in the $CePd_3$ lattice.[8] We estimate ω_{PM} to be about 85 meV. The fitted strength is about right, at 43 meV, but the phonon mode does not correctly reproduce the asymmetry. Alternatively, we observe that a sharp drop in A can be produced by a sudden increase in the optical conductivity σ at an energy E_s. In the simplest case we modify the low-frequency conductivity σ_n by a step function

$$\text{Re}\sigma(\omega) = \text{Re}\sigma_n(\omega)[1 + f\theta(E_s - \hbar\omega)], \tag{6}$$

where f is the fractional increase in the conductivity at the onset. The nature of the channel opening up in this model is unknown.

Shown in Fig. 2 is our best fit to the low-temperature data for $CePd_3$ using a step in the conductivity superimposed on the main resonance in $(\omega_p{}^2\tau)_e$. The fit assumes a 40% increase in the conductivity at 14.8 meV. The fit is excellent up to the maximum at 19 meV, especially in the

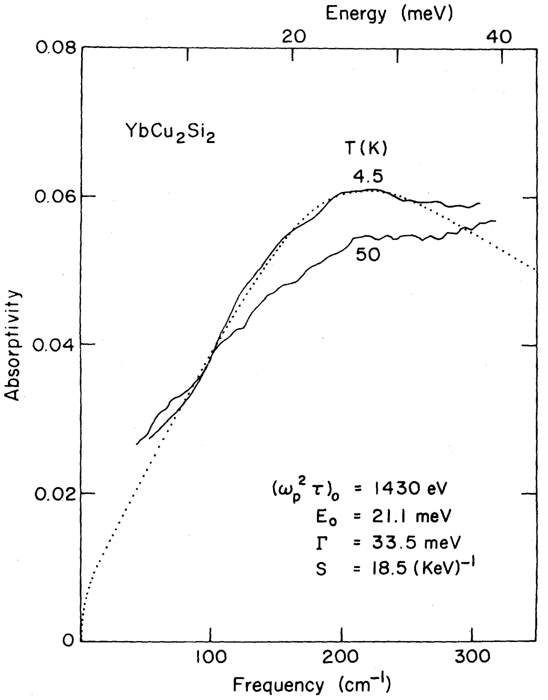

FIG. 3. Absorptivity vs frequency for $YbCu_2Si_2$. The resolution is 1 meV. The dotted curve is a fit to the 4.5-K data with use of the resonant-level model [Eq. (5)] with the listed parameters.

region of positive curvature below 15 meV. Above 19 meV, however, the resonance comes back down faster than the data. The difference, also shown, could represent a sideband of the resonance at 17.6 meV, or perhaps a second resonant level at higher energy.

Our optical results can be compared with the inelastic neutron scattering measurements on $CePd_3$ by Holland-Moritz *et al.*[9] Although those authors do not report an inelastic peak near 20 meV corresponding to the optical anomaly, their lowest-temperature measurements were at 100 K, which is just where the optical anomaly vanishes. There is no structure in the optical data similar to the quasielastic peak with a temperature-independent 19-meV width reported in the neutron work.

The frequency and temperature dependence of the absorptivity of $YbCu_2Si_2$ is very similar to that of $CePd_3$, as shown in Fig. 3. The 4.5 K data are fitted well by a resonance in $(\omega_p{}^2\tau)_e$ with $E_0 = 21.1$ meV and $\Gamma = 33.5$ meV, as shown by the dotted curve in Fig. 3. There is no sharp secondary feature in this compound corresponding to the one in $CePd_3$. No structure is seen in the analog integral-valent compound $LuCu_2Si$.

Table I summarizes our experimental results. The broad structure in the dielectric function of $CePd_3$ and $YbCu_2Si_2$ occurs at an energy comparable to the characteristic valence-fluctuation temperature kT_χ inferred from the temperature dependence of the magnetic susceptibility and to the temperature kT_ρ of the maximum in the dc electrical resistivity. The order of magnitude difference in kT_χ and kT_ρ for $CeAl_3$ and $CeCu_2Si_2$ compared with $CePd_3$ and $YbCu_2Si_2$ suggests that the absence of a resonance above 4 meV in the former two compounds is to be expected. All of these results are consistent with the hypothesis

TABLE I. Summary of our results on valence-fluctuation compounds. Listed are the parameters of the resonant-level model [Eq. (5)], along with energy scales in these materials inferred from magnetic susceptibility and electrical resistivity measurements.

Compound	E_0 (meV)	Γ (meV)	kT_χ (meV)	kT_ρ [a] (meV)
$CePd_3$	17.6	7.9	25 [b]	10
$YbCu_2Si_2$	21.1	33.5	9–24 [c,d]	17 [e]
$CeCu_2Si_2$	no resonance above 4 meV		1 [f]	3.1
$CeAl_3$	no resonance above 4 meV		~1 [g]	2.6

[a] This work.
[b] Ref. 10.
[c] Ref. 11.
[d] $YbCu_2Si_2$ is anisotropic.
[e] Corrected for the resistivity of isostructural nonmagnetic $LuCu_2Si_2$ compound.
[f] Ref. 12.
[g] Ref. 13.

that a broad optical feature in the far infrared, due to electron scattering from a resonant level, is characteristic of valence-fluctuation materials.

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