Intermediate-valent SmB_6 and the hybridization model: An optical study

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Reflectivity measurements in the far-infrared region at 4 K show that the SmB_6 spectrum is semiconductorlike with a reflectivity maximum located at 5.5 meV. This peak is absent at 300 K and the spectrum is metal-like. Optical constants are calculated by means of the Kramers-Kronig relations; the dielectric constants are unusually large and yield at 4 K a gap value of 3—4.7 meV. A two-oscillator fit of ϵ_2 has been calculated and the evolving oscillator strength has been used to evaluate in first approximation the interband density of states: The results are compared to the predictions of the hybridization model.

INTRODUCTION

Intermediate valence (IV) is the field in modern solid-state physics where even after 13 years of research experimental physics is much more advanced than theoretical explanations. The main reason for this is that theory is not yet sufficiently developed to treat the many-particle interactions of highly correlated electrons. Renormalization theories[1,2] seem to be the most promising, but most proposals for the explanation of experimental results start from simplified models. One of the first ideas was that the degeneracy of a $4f^n$ and $4f^{n-1}5d$ state at the Fermi level E_F would cause a hybridization of f and d states with an interaction energy Δ of order 10 meV.[3] One of the models treated is shown in Fig. 1(a),[4,5] where a broad one-electron band of $5d$ character is assumed to be degenerate with a $4f$ state. The $4f$ state is given a weak dispersion, and this model is thought to be typical for most Ce ($CeAl_3$, $CePd_3$, etc.) or Yb (YbCuAl, etc.) alloys. In these materials it is possible that a weak direct $4f$ overlap exists. On the other hand, crystal-field, electron-phonon, or other interactions can be assumed to cause a certain width (≈ 10

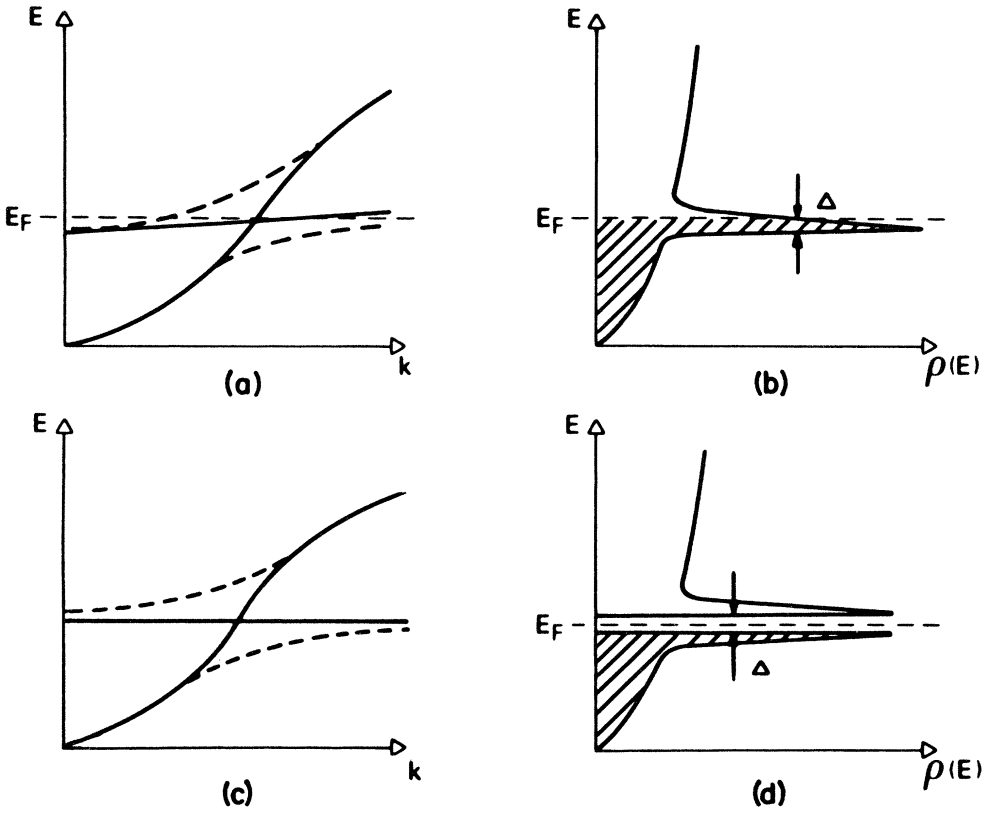

FIG. 1. Energy dispersion and density of states curves for two hybridization models.

meV) of the $4f$ states. We do not have to go into the details of possible mechanisms to realize that the density of states $\rho(E)$ will look like in Fig. 1(b) with a very large peak near E_F having a width of about Δ. Experimental evidence for such a model is indeed given by the extremely large γ values of the specific heat of up to 1600 mJ/mole K^2 for these materials,[6] more than 2000 times larger than for other metals (Cu, 0.5 mJ/mole K^2). The γ-term measures the density of states at E_F. Optical measurements[7] and point-contact spectroscopy[8] also indicate such a density of states peak at E_F.

If on the other hand the $4f$ state is taken to be dispersionless as indicated in Fig. 1(c), the repulsion of the wave functions at the intersection should lead to a gap in the density of states with E_F at $T\rightarrow 0$ being somewhere in the gap [Fig. 1(d)].

We are well aware of the difficulties of the hybridization model, pointed out by Anderson,[9] who argued that only one of the degenerate $4f$ wave functions can hybridize with the $5d$ states, and thus a gap should not form. On the other hand, Mott[10] has put forward the argument that it is exactly the high correlation of the $4f$ electrons which enforces a hybridization of all wave functions if one is tending to hybridize. In any case, gaps in the order of a few meV are an experimental fact in compounds such as SmB_6, "gold" SmS, and TmSe. The first evidence for a gap in SmB_6 appeared in the resistance increase at low temperatures,[11] but conclusive evidence has only come from a rigorous analysis of the temperature dependence of the Hall effect,[12] which gave an activated carrier concentration and only 5×10^{17} e/cm^3 at 4.2 K. In SmB_6 tunnel spectroscopy[13] and optical transmission[14] in the far infrared also revealed a gap. The most inpressive progress has been achieved with point-contact spectroscopy[15] and GaAs tunneling spectroscopy[16] because they are universally applicable and directly yield gap widths.

In SmB_6 (Refs. 11 and 17) and "gold" SmS,[18] however, large γ terms of the specific heat have been reported nevertheless, indicating high density of states at E_F, thus contradicting the gap model. Recently it was shown, however, for SmB_6 (Ref. 14), by extending the specific-heat measurements to lower temperatures than before and detecting additional impurity-dependent structures, that a linear term in the specific heat cannot be defined, making the above argument invalid. A similar, revised measurement on "gold" SmS still is necessary. In SmB_6, thus, every measurement is compatible with a gap and E_F being in the gap. However, there is not the slightest experimental evidence that two large peaks in the density of states are above and below E_F as shown in Fig. 1(d). It is the purpose of this paper to show by detailed optical analysis, that these density of states peaks really exist.

REFLECTIVITY MEASUREMENTS

The optical reflectivity of a large ($5\times 3\times 2$ mm^3) SmB_6 single crystal has been measured over the photon-energy range (1.5 meV—8 eV) in the temperature region between 4 K and room temperature with the use of three spectrometers: In the far-infrared region we have used a Bruker-Fourier spectrophotometer. The crystal is the same on which the Raman effect[19] and the point-contact spectroscopy[15] has been performed. The reflectivity results are shown in Fig. 2; at room temperature the spectrum is metal-like with a small shoulder at 0.1 eV and a screened plasma edge at 1.75 eV. Interesting is the reflectivity behavior at low temperatures, e.g., at 4 K. The reflectivity is reduced by 10% in the far-infrared region and a new maximum appears at about 5.5 meV, far below any observed optical-phonon frequency.[19] The reflectivity reaches the constant value of 90% for ω towards 10^{-3} eV. Below 10^{-4} eV and towards $\omega\rightarrow 0$ the reflectivity has been extrapolated to 100% to take into account a finite conductivity at 4 K.[12–14] This spectrum is semiconductorlike although it is the first time that a semiconductor with such a high reflectivity at 4 K has been observed. In Fig. 2 the dot-dashed curve is the $\omega\rightarrow 0$ extrapolation and the dotted curve gives the upper limit of the experimental error for the 4 K curve (solid curve). A detailed study of the temperature dependence of the reflectivity has revealed that the 4 K spectrum turns into the room-temperature spectrum already somewhat above 10 K. The reflectivity measurement at room temperature is in good agreement with results of[20] obtained for photon energies above 30 meV.

KRAMERS-KRONIG TRANSFORMATIONS

The reflectivity spectrum of SmB_6 has been analyzed in terms of optical constants by means of the Kramers-Kronig relations; a conventional extrapolation of the reflectivity above 8 eV has been made.[21] Figure 3 shows the real (ϵ_1) and the imaginary (ϵ_2) part of the dielectric constant for $T=4$ and 300 K. (The dotted curves at 4 K in Fig. 3 would be obtained using the dotted reflectivity curve of Fig. 2.) We note immediately that we are dealing with a very unusual material. First of all, at 4 K the dielectric constants are about 100 times larger than those of a normal semiconductor. The dc part of ϵ_1 falls be-

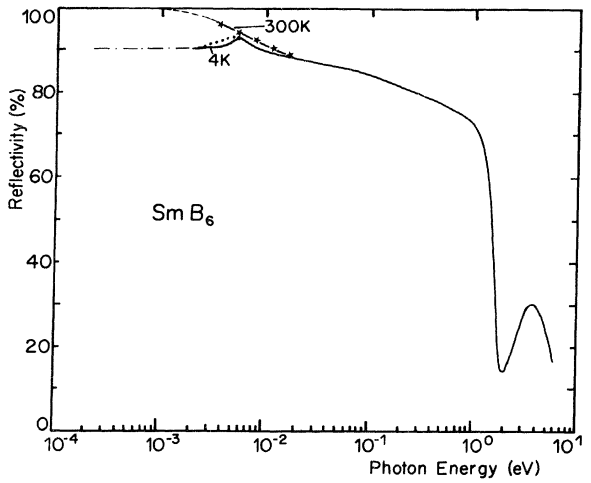

FIG. 2. Reflectivity of SmB_6 at 300 and 4 K. For the low-temperature curve the dot-dashed line is the $\omega\rightarrow 0$ extrapolation, and the solid and the dotted lines are the lower and upper limit of error of the far-infrared measurements.

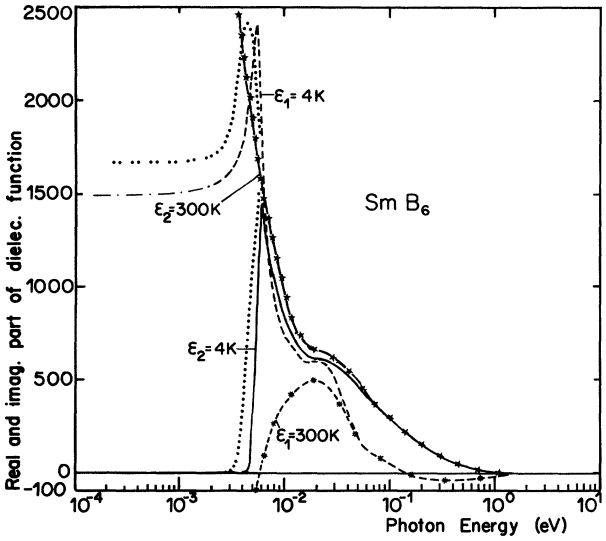

FIG. 3. Real (ϵ_1) and imaginary (ϵ_2) part of the dielectric functions of SmB_6. Punctuation is the same as in Fig. 2.

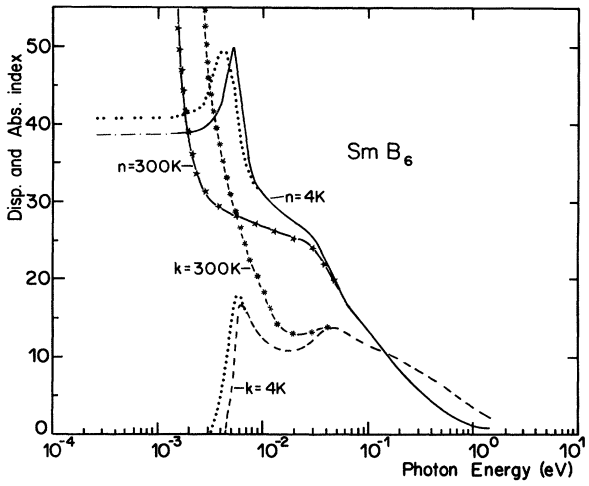

FIG. 4. Dispersive index n and extinction coefficient k of SmB_6 in the resonance region at 300 and 4 K.

tween 1500 and 1700 and reaches the value of 2000 in the resonance region. This implies a refractive index n of 40 to 50 shown in Fig. 4. Second, the small (10%) change in the reflectivity between 4 and 300 K in the far infrared has drastic consequences in terms of the dielectric constants; for example, ϵ_1 for $\omega \approx 10^{-4}$ eV amounts to 1600 at 4 K and drops to very large negative values at 300 K. Observing the imaginary part ϵ_2 at 4 K we note two absorption maxima, the first, very sharp one, located at 5.5 meV with an absorption edge between 3 and 4.7 meV, and the second, weaker one, centered at 0.1 eV, having instead a 100 times larger width. At 300 K ϵ_2 still exhibits the peak at 0.1 eV but diverges positively.

In order to assign the electronic transitions corresponding to these two peaks at 4 K we must consider in zeroth approximation the hybridization model; in this crude model the energy dispersion of the valence [hatched region of Fig. 1(d)] and of the conduction band will have f and d character at the Brillouin-zone boundary, respectively, and d and f character at the zone center, respectively: Valence and conduction band are then separated by the hybridization gap.

The very-lowest-energy transition with absorption between 3 and 4.7 meV will thus have f-f character at the onset of the transition (parity forbidden for direct optical transitions), the second transition will have a d-f matrix element. The absorption constant K for the meV transition at 4 K amounts to 2×10^4 cm^{-1} at the maximum, very unusual for the expected indirect transition [Fig. 1(c)]: Normally one would expect for this kind of transition a K value of the 10—30 cm^{-1}. This anomalously large K value of SmB_6 in the far-infrared region on the one hand is due to increasing f-d mixing for energies near the maximum of the first transition, and on the other hand it is due to the giant density of states at the Γ point for the conduction band and at the zone boundary for the valence band.

For $k_BT \gg \Delta$, e.g., at 300 K, gaps are no longer detectable experimentally because most electrons are thermally excited above the gap and the materials behave like metals. In Fig. 3 this is indicated by the large negative values of ϵ_1 at 300 K for $\omega \to 0$.

A collective oscillation of carriers with a plasma resonance is obtained whenever $\epsilon_1 = 0$ and $(d\epsilon_1/d\omega)_{\epsilon_1=0} > 0$. We notice that, at 300 K, ϵ_1 in Fig. 3 has three zero crossings, where those at 1.75 and 0.0053 eV fulfill the conditions for a longitudinal oscillation of free carriers. The transition at about 0.1 eV with $(d\epsilon_1/d\omega)_{\epsilon_1=0} < 0$ indicates a transverse excitation and is due to the d-f interband transition.

The two plasma resonances correspond to light (d) and heavy (f) electrons, which generally must be assumed to give rise to coupled oscillations. However, since the resonances are so far apart the coupled system has its eigenfrequencies practically at the resonance frequencies of the uncoupled resonators. In this case we can compute the independent plasma frequencies, which are screened by ϵ_{opt} of higher-energy interband transitions. For the resonance at 1.75 eV we use $\epsilon_{\text{opt}} \approx 2.7$ due to p-d interband transitions located around 4 eV (Fig. 2) and from $\omega_{pd}^2 = (4\pi e N_d)/(m_d^* \epsilon_{\text{opt}})$ obtain $(N_d m)/m_d^* = 0.42$ electrons per Sm ion. From the resonance at 0.0053 eV we use $\epsilon_{\text{opt}} \approx 400$ due to d-f interband transitions located at 0.1 eV (Fig. 3) and from $\omega_{pf}^2 = (4\pi e N_f)/(m_f^* \epsilon_{\text{opt}})$ obtain $(N_f m)/(m_f^* = 5.7\times10^{-4}$.

The valence determination of Sm in SmB_6 by means of the isomer shift of the Mössbauer effect and the lattice parameter[22,23] yields a practically temperature-independent valence of 2.6. We thus expect $N_f \approx 0.4$ and $N_f + N_d = 1$. Using these numbers, we obtain $m_d^* \approx 1.4(\pm 0.1)$ and $m_f^* \approx 1000m(\pm 500)$. Optical effective masses m_d^* of this magnitude have been observed before for $5d$ electrons in the La and Gd chalcogenides.[21] An optical effective mass m_f^* of about $1000m$ has also been inferred from optical measurements on "gold" SmS.[24]

Figure 5 shows the effect of the hybridization gap on the real part σ_1 of the optical conductivity. At 4 K the extrapolated dc value of σ_1 is about 2×10^{11} Hz (0.22 Ω^{-1} cm^{-1}), and above the transition threshold ener-

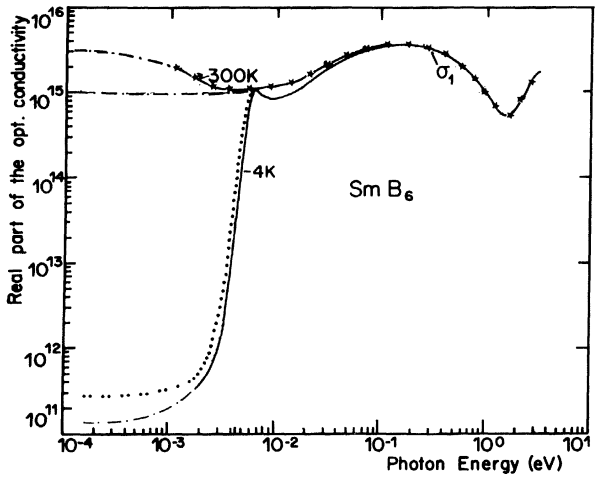

FIG. 5. Real part (σ_1) of the optical conductivity of SmB_6: Note the large difference between the dc value at 4 and 300 K.

gy it increases by 4 orders of magnitude (10^{15} Hz). At room temperature the extrapolated dc value of σ_1 is between 10^{15} and 3×10^{15} Hz, and thus metal-like. Our dc σ_1 values at 4 and 300 K are confirmed by electrical conductivity measurements,[13] although there is some sample-dependent scattering in these numbers. At 0.1 eV the optical conductivity increases to about 6×10^{15} Hz due to the *d-f* transitions; this latter behavior has been observed already in preceding papers.[13,20]

In Fig. 6 is given the number of electrons per Sm ion connected with the *f-f* and *d-f* transitions. The curve exhibits a first saturation plateau at about 1 eV with 1 effective electron per Sm ion. The above results indicate that the *f-f* and *d-f* transitions are exhausted for energies up to about 1.5 eV. At 4 K the effective number of electrons with mainly *f* character, i.e., up to energies of about 10^{-2} eV, is only about 10^{-3} instead of the expected 0.4. However, if we consider all transitions starting with the filled *f* states, we realize that the *f-f* transitions are only a small fraction of the total.

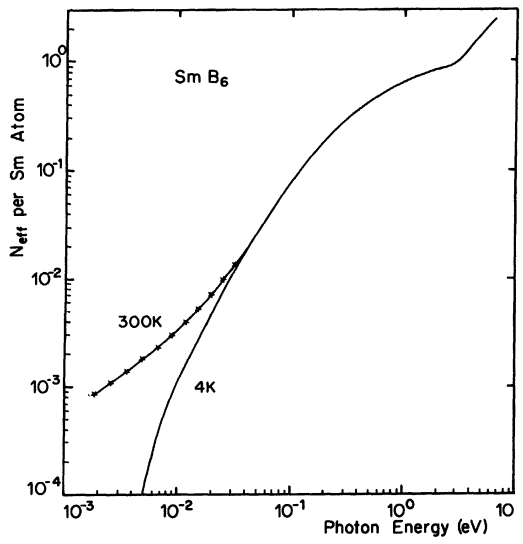

FIG. 6. Effective number of electrons per Sm ion connected with interband transitions.

TABLE I. Input parameters of the ϵ_2 fit.

	$\hbar\omega$ (eV)	$\hbar\Gamma$ (eV)	*f*
Oscillator 1	0.0055	0.0016	0.00033
Oscillator 2	0.1	0.5	0.3

DISCUSSION

To understand quantitatively these two electronic transitions we have tried to fit ϵ_2 in the near- and far-infrared regions with a simple two-oscillator function

$$\epsilon_2=\frac{4\pi e^2N}{m}\sum_{i=1}^{2}f_i\frac{\omega\Gamma_i}{(\omega_i^2-\omega^2)^2+\Gamma_i^2\omega^2},$$

where f_i is the oscillator strength, Γ_i is the damping constant, and ω_i is the resonance frequency for the *i*th oscillator: In a first approximation we assume only direct optical transitions. The input parameters of the ϵ_2 fit are summarized in Table I. The result of the fit is depicted in Fig. 7; the solid line is the calculated ϵ_2 curve, and the dotted line is ϵ_2 as obtained from the Kramers-Kronig analysis of the reflectivity.

How can we interpret the oscillator strength? For the low-energy resonator the oscillator strength is only 0.000 33, which implies that the transition is nearly forbidden. This fact corroborates the mainly *f-f* character of the first transition as mentioned above. For the higher-energy oscillator the transition probability is 0.3: *d-f* transitions are allowed from parity selection rules and responsible for this large matrix element. These results agree perfectly with the *f-d* hybridization model, which can explain exactly this kind of transition as a consequence of the electronic energy dispersions arising from the Sm $4f^6(^7F_0)$-$4f^5(^6H_{5/2})5d$ mixing.

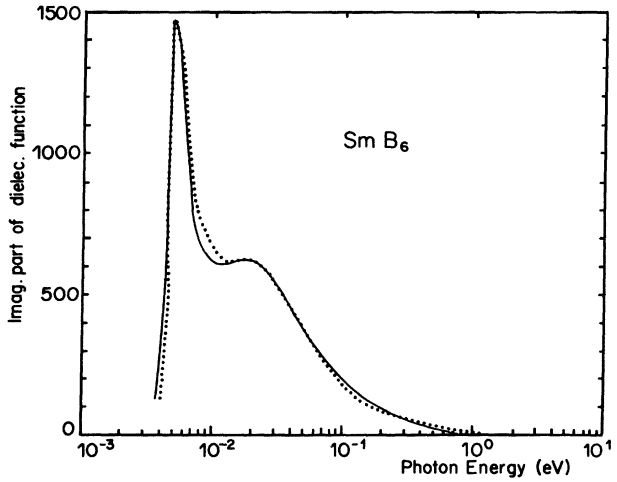

FIG. 7. ϵ_2 fit: Dotted line is ϵ_2 at 4 K from Fig. 3 and solid line is the fit result.

At this point we can estimate in first approximation the joint density of states J_{cv} assuming constant matrix elements within an f-f and d-f transition, respectively. We can describe the imaginary part of ϵ_2 in terms of an integration over a surface in the reciprocal space k. For direct optical transition one finds

$$\epsilon_{2_i}(\omega)=\frac{e^2}{\pi m^2\omega^2}\int |a_0 p_{cv}|_i^2 \frac{dS}{|\nabla_k E_{cv}(k)|_i}$$

$$=8\left[\frac{\pi e}{m\omega}\right]^2 |a_0 p_{cv}|_i^2 J_{cv_i} ,$$

where dS is a simple surface element in k space such that $E_{cv}(k)=\hbar\omega$, and p_{cv} is the momentum operator. The oscillator strength is coupled to p_{cv} with a relation

$$f_i=2|a_0 p_{cv}|_i^2/\hbar\omega m .$$

It is clear that in this approximation we can calculate from ϵ_2 the joint density of states J_{cv}: $2J_{cv}$ is equal to the interband density of states (IDS). The IDS arising from this calculation is shown in Fig. 8: There exists a giant peak of about 900 states per Rydberg per cell located at 6.3 meV and a shoulder of about 4 states per Rydberg per cell at 0.1 eV. The estimated error in this approximation is roughly a factor of 2. If we had explicitly taken an indirect f-f transition into account, the matrix elements would have become smaller, and thus J_{cv} would have become even larger. Our value of J_{cv} for the f peaks is thus only a lower limit. The hybridization model provides for SmB_6 a large density of states with two sharp, separated maxima at E_F and a weaker maximum for higher-excitation energy [Fig. 1(d)]; therefore the IDS, which is a folding of occupied and empty states, must also have a giant peak in the meV region and a weaker but broader maximum at higher energies; we can now state that the curve in Fig. 8 corresponds to the hybridization model discussed above.

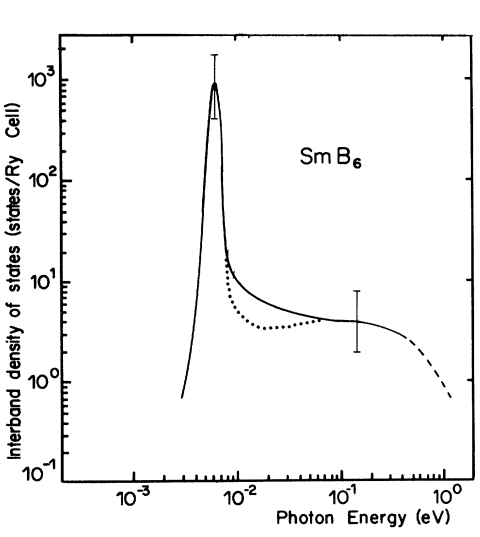

FIG. 8. Interband density of states (IDS) of SmB_6 at 4 K calculated with the use of the resulting fit oscillator strength in the constant matrix element approximation: Dotted line gives the uncertainty between peak with f character and the one with d character.

The optical density of states with f character has a maximum at 6.3 meV. Usually this energy is the difference between the maxima of the density of states of conduction and valence bands. We assign the shoulder at 0.1 eV to the optical band with d character, which amounts to 4 states per Rydberg per cell: a similar density of states must also be present in LaB_6. Walch *et al.*[25] found in their calculation 5.01 states per Rydberg for LaB_6 and 6.05 for YB_6. Within the estimated error, our results agree very well with this calculation for the band with d character. The halfwidth of the IDS at the low-energy transition is 5 meV, which means that this transition occurs between two atomiclike levels separated by Δ. In other words, the density of states of the occupied and empty "f bands" is so much larger (about 300 times) than the one of any other band that we are in effect dealing with a "two-level" system. The total width of the f bands is about 10 meV, and thus much smaller than the one assumed by Takigawa[26] (40 meV).

Specific heat on SmB_6 has been measured recently again[14]: The paper reports the difference $C_{\text{sample}}-C_{\text{lattice}}$ as function of the temperature. At 10 K the specific heat is about zero; for $T>10$ K it increases until it reaches a maximum value at $T_{\max}=32$ K and diminishes slowly at higher temperatures. This behavior can be interpreted within an electronic two-level system as a consequence of the Boltzmann distribution. We can then interpret this specific-heat maximum with a Schottky anomaly. A Schottky anomaly yields the following relation:

$$0.41=k_B T_{\max}/\epsilon ,$$

where ϵ is the energy between the two levels: it follows that $\epsilon=6.7$ meV. For SmB_6 this value corresponds not to the gap energy but to the energy difference between the two maxima of the conduction and valence density of states: This value corresponds very well to our calculated value of 6.3 meV.

CONCLUSION

The far-infrared reflectivity measurements at 4 K and the determination of the optical constants corroborates the much discussed model of the hybridization gap for intermediate-valent SmB_6. We have interpreted the low-energy maximum in the reflectivity and the absorption edge at 3—4.7 meV in ϵ_2 as being due to electronic transitions with mainly f character across the hybridization energy gap: The gap value of 4.7 meV agrees very well with the one obtained from point-contact spectroscopy.[15] With this contribution we wanted to show that the hybridization model resulting in a gap and two large density of states peaks above and below E_F has a large probability for SmB_6.

Many questions, however, are still unsolved. How does

the hybridization mechanism work for degenerate f states? How can such a small gap exist in all directions? Why is it that imperfections, which must be present up to 1%, do not smear the gap? Normally defects would make this gap appear only locally. However, a local gap would not show up in the reflectivity $R \to 1$ for $\omega \to 0$, and consequently ϵ_1 and ϵ_2 would diverge towards $-\infty$ and $+\infty$, respectively, as in a metal. A possible explanation may be found in impurity screening: We have seen that this material has a very high dielectric constant of about 1500—2000. This extremely high value would drastically reduce the impurity disturbance. It appears that the f-d hybridization and the small width of the gap must have to do with a coherent effect. One should find such a parameter which expresses this coherency and which diverges for $k_B T < \Delta$ and gets reduced as the temperature increases: Above a critical temperature it will be zero or it will merge into a fluctuating regime; in this last case the hybridization gap would become local and it would disappear when $k_B T \gg \Delta$. When the hybridized compounds for $k_B T \ll \Delta$ behave similarly to two-level systems, it should also be possible to derive the magnetic susceptibility for such a system. However, in contrast to the specific heat, quantum numbers are now necessary and it appears as a formidable task.

From the experimental point of view TmSe is also very interesting, because it poses the question of whether the experimentally observed gap is a hybridization gap or an antiferromagnetic gap. Similar optical experiments are much more difficult since the gap is about 2 meV (Ref. 15) much less than in SmB_6, which probes the limits of Fourier spectrometers, and the temperature for observation must be less than 3 K, since $T_N \approx 3$ K.

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