Dielectric Response of SmB_6 in the Millimeter Wave Range

M. DRESSEL[1]) (a), B.P. GORSHUNOV (a,b), N.E. SLUCHANKO (b), A.A. VOLKOV (b), B. HENDERSEN (c), G. GRÜNER (c), G. KNEBEL (d), A. LOIDL (d), and S. KUNII (e)

(a) 1. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Germany

(b) General Physics Institute, Russian Academy of Sciences, 117942 Moscow, Russia

(c) Department of Physics, University of California, Los Angeles, California 90095, USA

(d) Experimentalphysik V, Universität Augsburg, D-86135 Augsburg, Germany

(e) Department of Physics, Tohoku University, Sendai 980, Japan

The low-temperature measurements of the conductivity and dielectric permittivity of single crystalline SmB_6 in the microwave and submillimeter spectral range give evidence for a 19 meV energy gap in the density of states and for a narrow donor-type band lying 3 meV below the bottom of the upper conduction band. It is shown that at temperatures 8 K < T < 20 K the dc conductivity and the electrodynamic response of SmB_6 in the frequency range up to the far infrared are determined by quasi-free carriers thermally excited in the conduction band; below 8 K the carriers are localized in the narrow band showing the typical signature of hopping conductivity.

Up to now the origin of the ground state of Kondo-insulating systems is not completely understood, though extensive experimental material has been accumulated and various theories have been proposed [1]. Of particular interest is the energy scale of several milli-electronvolts because in this range the hybridization gap appears in the density-of-states (DOS) spectra at low temperatures. In this report we present the results on direct measurements of the microwave, millimeter and submillimeter optical response of samarium hexaboride SmB_6 which is sometimes considered to be the prime representative of the family of Kondo insulators. The magnetic susceptibility of SmB_6 reveals the main features of an intermediate valence compound: a Curie-Weiss like susceptibility for $T > 100$ K indicating local-moment behavior and a Pauli spin susceptibility for low temperatures due to a non-magnetic configuration for $T \to 0$ K. The cusp in the susceptibility close to 80 K can be taken as a rough estimate of the characteristic spin-fluctuation temperature T^*. In Kondo insulators the gap due to the coherent on-site hybridization between the narrow 4f states with the band states is expected to be of the same order of magnitude. There is no general agreement on the value of the gap in SmB_6. Depending on the measurement techniques used, a rather broad distribution of gap values is reported which may be divided into two groups: small (3 to 5 meV) and large (10 to 16 meV) gaps (for reviews see [1, 2]). One of the most straightforward

[1]) Corresponding author: Fax: +49-711-685-4886; e-mail: dressel@pi1.physik.uni-stuttgart.de

methods to measure a gap in the DOS is provided by the optical spectroscopy. However, in Kondo-insulators such task is nontrivial since the smallness of the energy scale makes it difficult to use standard far infrared techniques. Thus we have applied submillimeter (submm) quasioptical spectroscopy which enabled us to precisely measure the conductivity $\sigma(\nu)$ and dielectric permittivity $\varepsilon(\nu)$ of SmB_6 in the energy range from 0.6 to 4.5 meV. To further investigate the origin of the low-temperature transport in SmB_6 we have also performed measurements of the microwave conductivity for the first time.

The single crystals of SmB_6 were grown by a floating zone method. Their high quality is characterized by a large dc resistivity ratio $\varrho(60\text{ mK})/\varrho(300\text{ K}) \approx 4 \times 10^6$. The submm spectra of conductivity and dielectric permittivity were measured *directly* (no Kramers-Kronig analysis used) in the range $\nu = 5$ to 36 cm^{-1} ($h\nu = 0.6$ to 4.5 meV) at $T = 3$ to 20 K by a transmission technique described in [3]. The infrared (IR) reflection was also measured, finally experiments on the dc resistivity and the Hall effect have been performed. In this paper we also present our results on the microwave (35 GHz) conductivity of SmB_6 measured by the cavity perturbation technique [4]. All measurements were done on samples cut of the same single crystal.

Our main findings are summarized in Fig. 1 where several characteristic types of behavior can be distinguished. Firstly, above 8 K the conductivity and permittivity below the FIR ($\leq 40\text{ cm}^{-1}$) display a typical Drude-like behavior which is demonstrated by the 13 K, 16 K, and 18 K spectra in Fig. 1: towards low frequencies the dielectric permittivity decreases, and the conductivity increases saturating to the dc values (as shown by dashed lines representing the fit, see below); our microwave results (squares in Fig.1a) confirm this behavior. Secondly, at temperatures below 8 K the $\sigma(\nu)$ dependence is no longer Drude-like since the submm and the 35 GHz conductivities here are about two orders of magnitude larger than the dc conductivity as seen in the 3 K spectrum. Thirdly, an absorption peak at about 24 cm^{-1} is clearly detected in the submm spectra. Finally, a semiconductor-gap-like increase of the FIR conductivity by more than an order of magnitude is seen around 100 cm^{-1}.

Accordingly, we have fitted the broad-band spectra with a model containing a Drude and an oscillator term (short-dashed lines in Fig. 1) which allowed us to extract microscopic parameters of the free charge carriers determining the low-frequency submm-microwave and the dc responses of SmB_6 at $8\text{ K} < T < 20\text{ K}$ [5]. The obtained temperature dependence of the squared plasma frequency ν_{pl}^2 and correspondingly of the charge carrier concentration $N = \nu_{\text{pl}}^2 m^* \pi / e^2$ (m^* is the effective mass, e stands for the electronic charge) reveals two regimes of activated behavior (see inset in Fig. 1a) indicating the presence of an energy gap of (19 ± 2) meV (line 1) in the DOS and a narrow donor-like band lying 3 meV (line 2) below the upper edge (see inset in Fig. 1b). At exactly the frequency (24 cm^{-1}) corresponding to the energy 3 meV we observe an absorption peak in the submm spectra which therefore can be associated with the direct photon-assisted excitations of carriers from this narrow band into the conduction band. These results account for the large discrepancy in the gap values reported in the literature ranging from 2 to 16 meV. The values of only a few meV correspond to the energy difference between the bottom of the conduction band and the peak in the DOS (narrow band) *inside* the hybridization gap. The latter one has a considerably larger width of $E_{\text{g}} = 19$ meV and corresponds to features reported in the range of 14 to 16 meV. Calculating [6, 7] the hybridization gap by $E_{\text{g}} \approx 2T^* \approx 160$ K yields a gap value of $160\text{ K} \approx 14$ meV in SmB_6 which is close to the experimentally observed gap

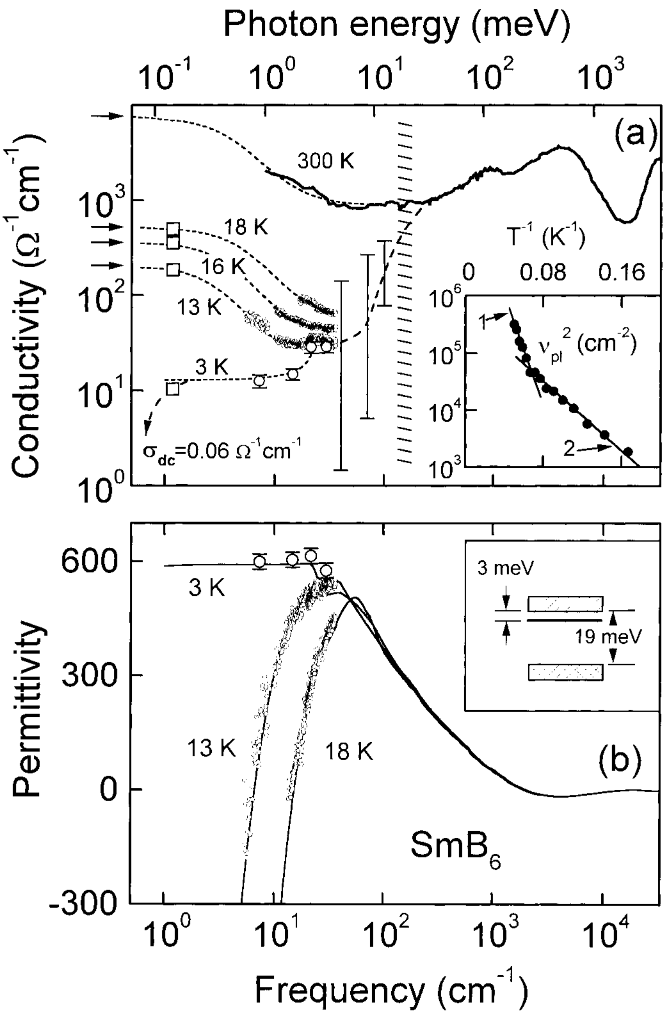

Fig. 1. Frequency dependence of a) the conductivity and b) dielectric permittivity of SmB_6. The open dots correspond to the submm data; the open squares to the microwave data at 35 GHz; the arrows to the dc conductivity. The solid lines are obtained by the Kramers-Kronig analysis of the FIR reflectivity measured in this work, combined with the spectra taken from [9] and normalized at lowest frequencies to the reflectivity calculated from submm ε and σ. The long-dashed lines interpolating the 3 K-data sets are guides to the eye. The large error bars for the IR conductivity correspond to a $\pm 0.25\%$ uncertainty of the 3 K reflectivity as obtained by the Kramers-Kronig analysis. The dashed lines show the result of the least square fit (see text and the data in [5]). The shaded area corresponds to the energy gap value (19 $\pm$ 2) meV. The inset in frame a) shows the temperature dependence of the squared plasma frequency with two types of activated behavior: $\nu_{pl} \propto \exp[(-19\ \mathrm{meV})/2k_BT]$, line 1, and $\nu_{pl} \propto \exp[(-3\ \mathrm{meV})/k_BT)]$, line 2 ($k_B$ is the Boltzman constant). The inset in frame b) gives a simplified view of the band scheme of SmB_6

energy. Concomitantly a spin gap of the same size is expected [7] and indeed a gap of 14 meV has been detected by neutron scattering [8].

Below $\approx$ 8 K the submm response in SmB_6 is no longer determined by the carriers in the conduction band, since they freeze out due to decrease of their concentration (inset in Fig. 1a). We assume that at these low temperatures the conductivity of SmB_6 is dominated by carriers moving within the narrow band. Here the dc conductivity is smaller compared to the submm conductivity and shows a typical Mott-like dependence on temperature $\sigma_{dc}(T) \propto \exp\,(T^{-1/4})$ while the submm conductivity is practically temperature and frequency independent [5]. Both behaviors indicate a hopping type of transport and, consequently, the existence of a characteristic frequency below which the response of quasi-free carriers is violated by the presence of localizing potentials leading to the $\sigma(\nu) \propto \nu^s$ dependence with $s \approx 0.8$ [10]. The data on the microwave conductivity presented here (Fig. 1a) indicate that this characteristic frequency is lying below 1 cm^{-1}. To get more details on low temperature electronic transport in SmB_6 lower-frequency experiments are in progress.

In conclusion, we have performed the first direct measurements of the dynamical conductivity and dielectric permittivity spectra of SmB_6 in the range 1 to 36 cm^{-1} at temperatures 3 to 20 K. The obtained results give evidence for a gap of E_g = 19 meV in the density of states and an additional narrow donor-type band lying 3 meV in the gap. It is shown that above 8 K electronic properties of SmB_6 are governed by free electrons thermally excited into the conduction band and below about 5 K by charge carriers moving within the narrow band.

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