# Dynamics of the coherent ground state in intermediate-valent $YbB_{12}$

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The optical properties of the intermediate valence semiconductor $YbB_{12}$ are studied at frequencies 8 $cm^{-1}$–10 000 $cm^{-1}$ (quantum energies 1 meV to 1.2 eV) and temperatures 5 K–300 K. Microscopic parameters of charge carriers are determined: effective mass, mobility, relaxation rate, relaxation time, plasma frequency. A strong decrease of the relaxation rate is observed when cooled down to 70 K, indicating that scattering is mediated mainly by phonons and not by the Kondo effect. Below the coherence temperature of $T^*=70$ K a Drude-like response of renormalized quasiparticles is observed with a scattering rate $\gamma$ following the Fermi liquid behavior $\gamma \propto T^2$, an effective mass $m^* \approx 34 m_0$, and an enhanced relaxation time $\tau = 4 \times 10^{-13}$ s, implying that coherent effects play a crucial role in formation of the ground state in $YbB_{12}$. At $T = 5$ K an absorption peak is found at 2.7 meV whose origin can be ascribed to an exciton-polaronic bound state.

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## I. INTRODUCTION

Intermediate-valence narrow-gap semiconductors belong to the class of strongly correlated electron systems for which the low-temperature properties are governed by coherent many body effects. In contrast to heavy-fermion compounds, they exhibit nonmetallic and typically nonmagnetic ground states due to narrow (of order of few meV) charge and spin gaps.[1,2] General aspects of the physics of intermediate-valence semiconductors are captured by theories based on a periodic Anderson Hamiltonian with a complete coherence of the Kondo sites and a Fermi-liquid like ground state.[2] However, some recent transport, thermodynamic, optical, and magnetic experimental results[3–11] have raised a number of questions about the nature of the ground state in semiconducting as well as in metallic correlated electron systems: Is the insulating gap connected with coherence at all? Is the ground state that of a true insulator or is there an intrinsic metallic phase at lowest temperatures? How crucial are valence fluctuations and magnetic and charge orderings for the formation of the ground state? To account for recent data, different models are put forward including those which do not incorporate coherence of the lattice or alternating valence as necessary elements[12,13] or viewing the development of a coherent state as partial condensation of Kondo centers into the lattice,[14] considering Wigner-crystal[12] or exciton-polaron[15] instabilities, predicting electron-type ferroelectricity due to coherence of the *d* and *f* states.[16]

The origin of the ground state of intermediate-valence semiconductors should be intimately connected with the mechanism of the charge-carrier dynamics. To study these issues with regard to a typical representative of intermediate-valence semiconductors $SmB_6$, we have recently applied broadband optical spectroscopy[17] with special emphasis to the lowest, THz and GHz frequencies where effective-mass and scattering-rate renormalization effects must be strongly pronounced.[2] Microscopic characteristics of heavy carriers (relaxation rate, effective mass, concentration, mobility) were obtained for $SmB_6$ and arguments were put forward in favor of exciton-polaron ground state.[15] In the present paper we discuss results of similar optical studies of another intermediate-valence semiconductor, $YbB_{12}$, performed at energies down to 1 meV. We were motivated by the fact that although $YbB_{12}$ in general reveals the typical behavior of intermediate-valence semiconductor, the standard *f*-*d* hybridization theory fails to account for some of its low temperature properties, like a complex gap structure,[18] anisotropic low-temperature magnetoresistance,[10,19] or independence of the gap value on doping with nonmagnetic Lu.[5,6,20,21]

$YbB_{12}$ has a cubic crystal structure ($Fm3m$) of NaCl type[22] with valence of Yb ions ranging from 2.86 to 2.95.[6,21,23,24] Transport, magnetic, and thermodynamic properties of $YbB_{12}$ demonstrate common features of an intermediate-valence semiconductor. Magnetic susceptibility shows a Curie-Weiss behavior for temperatures above 170 K and reveals[3] a maximum at around $T=75$ K indicating a nonmagnetic low-temperature phase with the magnetic gap of 10 meV.[20] Recent inelastic neutron scattering evidences short-range antiferromagnetic fluctuations with two sharp excitations at 14.5 and 20 meV.[11] From electronic specific heat and resistivity measurements a transport gap of 10–12 meV was identified.[25,26] These values coincide with the gap of 12 meV from photoemission[18] and electron spin resonance[27] (ESR) experiments. At $T<80$ K an optical gap of 20–25 meV develops producing a shoulder at 40 meV.[28] The value of 10 meV is believed to descend from a hybridization gap while the entire gap structure needs further detailed studies.[18,28–30] The latest optical data[31] on crystals with improved quality indicate a gap value of 15 meV associated by the authors with an indirect gap within the picture of a renormalized band model of a Kondo semiconductor; the feature at 0.2–0.25 eV is ascribed to a direct gap.

## II. EXPERIMENTAL DETAILS

The $YbB_{12}$ single crystals for the present study were grown as described in Ref. 32. The optical measurements utilized two spectrometers: a Bruker IFS 113V was used to measure the reflectivity from the far-infrared range up to frequencies $\nu=10\,000\ \text{cm}^{-1}$. By employing terahertz quasioptical technique[33] we succeeded in performing reliable reflectivity measurements down to frequency of $\nu=8\ \text{cm}^{-1}$ or even $6\ \text{cm}^{-1}$ (corresponding to photon energies of about 1 meV). To get the spectra of optical constants of $YbB_{12}$ we performed a Kramers-Kronig analysis of the merged reflectivity spectra, with the low-frequency Hagen-Rubens extrapolation according to the measured dc conductivity,[32,34,35] and for the high frequency extrapolation we utilized optical data previously taken on the same crystals.[28] In addition to the reflection experiments, direct measurements of the GHz and THz spectra of the dynamical conductivity and dielectric constant were performed at the lowest temperature of 5 K by measuring the complex transmissivity[36] of a thin (0.02 mm) sample.

### III. RESULTS AND ANALYSIS

Figure 1 presents the spectra of reflectivity $R(\nu)$, conductivity $\sigma(\nu)$, and dielectric constant $\epsilon(\nu)$ of $YbB_{12}$ at several temperatures. All features observed in previous optical experiments[28] are clearly seen: in particular, an "infrared peak" at 250 meV, a shoulder (or pseudogap like feature) at 40 meV in $\sigma(\nu)$ appearing below 70 K, and a pronounced, strongly temperature-dependent Drude-like component in the $\sigma(\nu)$ and $\epsilon(\nu)$ spectra along with the typical plasma edge in $R(\nu)$. We note that we do not see a bump at 15 meV recently reported[31] on high-quality crystals. This, however, does not affect the main subject of this paper: we report on the quantitative analysis of temperature behavior of free charge carriers that determine all electronic properties of $YbB_{12}$ which has become possible since we were able to measure the optical response of $YbB_{12}$ down to very low frequencies of about $8\ \text{cm}^{-1}$. In the analysis we performed a simultaneous least-square fit of the $R(\nu)$, $\sigma(\nu)$, and $\epsilon(\nu)$ spectra shown in Fig. 1. To model the free-carrier response a Drude expression for the complex conductivity[36] was used

$$\hat{\sigma}(\omega) = \sigma + i\frac{\omega}{4\pi}\epsilon = \frac{ne^2\tau}{m^*(1-i\omega\tau)}, \qquad (1)$$

where $n$ and $e$ are the concentration and the charge of the carriers, $\tau=1/(2\pi\gamma)$ is their relaxation time, $\gamma$ is the relaxation frequency, $m^*$ is the effective mass, $\omega=2\pi\nu$. In addition, Lorentzian terms were used to describe the 250 meV and the 40 meV features, as was done in Ref. 28. Assuming a single band with one type of carriers, we used the Hall data obtained on crystals from the same source[32] in order to evaluate the carrier concentration $n=1/(ecR_{\text{Hall}})$. This allowed us to calculate the temperature dependence of the plasma frequency $\nu_p=\omega_p/2\pi=(ne^2/\pi m^*)^{1/2}$, the effective mass $m^*=ne^2/(\nu_p)^2$ and the mobility $\mu=e/(2\pi m^*\gamma)$. The validity of the calculations is confirmed by the fact that the obtained mobility at all temperatures coincides well with the Hall mobility $\mu_{\text{Hall}}=\sigma_{\text{dc}}R_{\text{Hall}}$ obtained from independent dc-conductivity measurements on the same crystals.[32,34,35] In addition, the determined scattering rate and effective mass

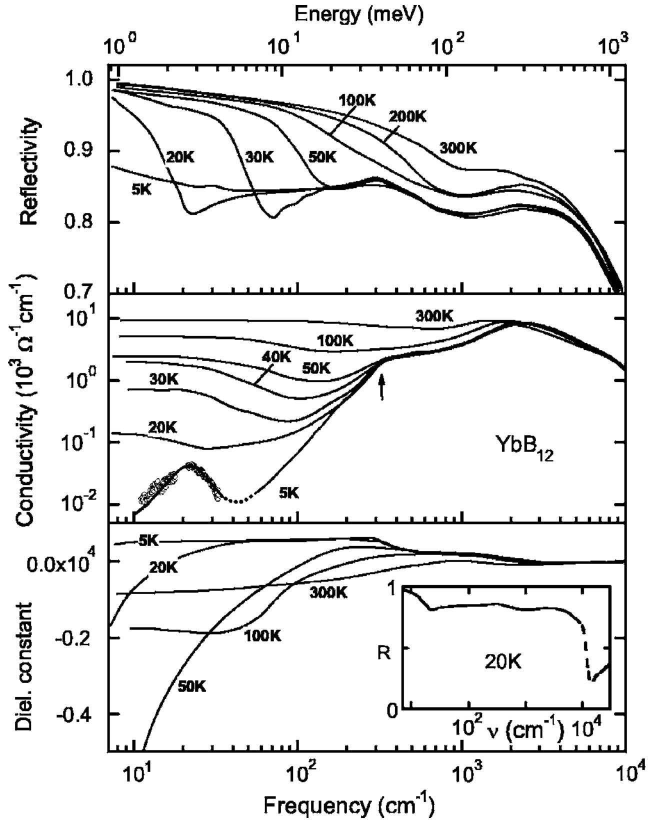


FIG. 1. Spectra of reflectivity, conductivity, and dielectric constant of $YbB_{12}$ at different temperatures as indicated. The absorption peak at $22\ \text{cm}^{-1}$ seen at $T=5$ K in the conductivity spectrum was recorded by measuring complex transmissivity of a thin (0.02 mm) sample; the solid line shows a Lorentzian fit with the eigenfrequency $\nu_0=22\ \text{cm}^{-1}$, dielectric contribution $\Delta\epsilon=75$, and damping $\gamma=15\ \text{cm}^{-1}$. Inset: broadband reflectivity of $YbB_{12}$ showing two plasma edges, at low (heavy quasiparticles) and high (unscreened electrons) frequencies. The dashed line corresponds to the data taken from Ref. 28.

agree well with the lowest frequency values of $\gamma(\omega)$ and $m^*(\omega)/m_b$ which we have calculated from the $\sigma(\nu)$ and $\epsilon(\nu)$ spectra within the generalized Drude approach,[36] after the interband transition at 250 meV has been subtracted from the spectra.

### IV. DISCUSSION

#### A. Heavy carrier dynamics

The obtained temperature dependences of the plasma frequency, the effective mass, the mobility, the scattering rate, and the relaxation time are presented in Fig. 2. We first consider temperatures above 100 K. While cooling from 300 K to 100 K the effective mass increases five times which can be associated with an increasing $f$ admixture in the joint quasiparticle density of states at the Fermi level, at temperatures close to the Kondo temperature $T_K\approx 220$ K in $YbB_{12}$.[37] The increase of the effective mass and an exponential decrease of the concentration (from the Hall data) lead to a strongly diminished plasma frequency (or spectral weight) of the carri-

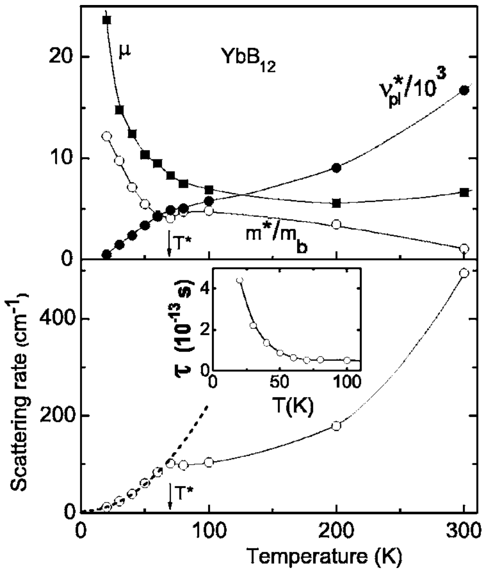


FIG. 2. Temperature dependences of charge-carrier parameters in $YbB_{12}$. Upper panel: mobility (in $cm^2\,V^{-1}\,s^{-1}$), effective mass $m^*/m_b$, plasma frequency (in $cm^{-1}$). Lower panel: scattering rate and relaxation time (inset). $T^*=70$ K indicates the coherence temperature.

ers. At these temperatures, a pronounced decrease of the scattering rate is clearly seen: when going from 300 K to 100 K the value of the scattering rate $\gamma$ decreases by a factor of five. This behavior is characteristic for a phonon-assisted scattering and contradicts a simple Kondo mechanism for which one would expect increasing strength of magnetic scattering upon cooling,[38] which also causes the mobility to decrease, while the latter is almost constant at these temperatures (Fig. 2). Also the temperature variation of resistivity of $YbB_{12}$, which shows an activated behavior rather than a Kondo-type dependence $\rho \propto -\ln T$, is in contrast to pure Kondo scattering. A possible reason for these observations is the presence of a gap in the density of states of $YbB_{12}$ at temperatures above 100 K. Indications of a gap at $T > 100$ K are also seen in photoemission experiments.[18]

At low temperatures, below $T \approx 70$ K, the gap at the Fermi level in the density of states opens in $YbB_{12}$ which produces a shoulder in the $\sigma(\nu)$ spectrum at 40 meV (Fig. 1) and leads to anomalies in transport, magnetic, and thermodynamic properties.[3,24,32] At the same temperature, an abrupt change in the charge carrier characteristics is observed (Fig. 2): the scattering rate starts to follow the dependence $\gamma(T) \propto T^2$ typical for a Fermi liquid,[39] and the effective mass begins to increase strongly. [Note, that the observed $\gamma(T) \propto T^2$ behavior does not depend on any assumption in our analysis of the experimental data.] These findings indicate that below $T^*$ a transition occurs into a state with full coherence among the $f$ sites[38,40] leading to the buildup of the heavy fermionlike quasiparticles. In other words, the low-frequency ($\nu < 100$ $cm^{-1}$) dispersion we observe in the spectra of $YbB_{12}$ at low temperatures ($T < T^* = 70$ K) is determined by a response of a Fermi liquid composed of heavy quasiparticles, $m^*(20\text{ K}) = 12 m_b$, with a strongly enhanced relaxation time, $\tau(20\text{ K}) = 4 \times 10^{-13}$ s (inset in Fig. 2) and mobility $\mu(20\text{ K}) = 24$ $cm^2/(V\,s)$. We then consider $T^* = 70$ K as a coherence temperature in $YbB_{12}$. Here $m_b$ is a band mass with the value of $m_b = 2.85 m_0$ obtained from the room temperature fit of experimental spectra ($m_0$ is the free electron mass).

Qualitatively, our observations resemble the optical response of heavy-fermion compounds where a Drude-like behavior of renormalized (heavy) quasiparticles is expected[39] and observed[2,41] in the coherent state, with a characteristic Fermi-like temperature-frequency variation of the scattering rate $\gamma \propto (2\pi k_B T)^2 + (h\omega)^2$. In case of $YbB_{12}$ we do not observe any frequency dependence of $\gamma$: according to the qualitative analysis given in Ref. 42 such dependence will not influence the optical conductivity if the relaxation time is much larger than the characteristic time $h/k_B T_K$ of electron transfer between neighboring lattice $f$ sites ($k_B$ is the Boltzmann constant and $h$ the Planck constant). Indeed, in our case of $YbB_{12}$ the relaxation time $\tau = 4 \times 10^{-13}$ s $\gg h/k_B T_K = 3 \times 10^{-14}$ s (with $T_K = 220$ K for $YbB_{12}$). Another feature of a heavy-fermion state is the presence of two plasma edges in the reflectivity spectra that originate from plasma oscillations of heavy (renormalized) and light (unrenormalized) quasiparticles. While usual plasma oscillations of unrenormalized charges normally lie in the region of a few eV, the renormalized plasmons are expected around the "screened" plasma frequency $\nu_p/\epsilon_{\rm inf}^{1/2}$ which can fall in the far-infrared or even microwave frequency range ($\epsilon_{\rm inf}$ is the high frequency dielectric constant). In $YbB_{12}$ the two correspondent plasma edges are clearly seen in the inset of Fig. 1.

### B. Excitonic-polaronic state

Let us now consider the lowest temperature spectra. As seen in Fig. 1, at $T = 5$ K the free carrier response is completely gone from the window of our measurements and a pronounced absorption peak is observed at 22 $cm^{-1}$ (corresponding to 2.7 meV). As is summarized in Ref. 15, the intermediate-valence semiconductors are inclined to an excitonic-like instability at low temperatures: due to the soft valence fluctuations with a characteristic frequency around $10^{13}$ to $10^{14}$ Hz, which is close to that of phonons, the moving carriers get self-trapped and form exciton-polaronic bound states within the gap. Signatures of an excitonic dielectric state have been observed in intermediate-valence systems, like $TmSe_{1-x}Te_x$,[43] and (Sm,Y)S.[15,44] Quite recently, ideas of excitonic dielectric state were successfully applied to account for low-temperature transport and optical properties in $SmB_6$, where an absorption peak was found at 24 $cm^{-1}$ and attributed to an exciton-polaronic bound state.[7,8,15,17] In analogy to $SmB_6$, we assume that the absorption at 22 $cm^{-1}$ in the low-temperature spectra of $YbB_{12}$ can be connected with formation of an excitonic band close to the bottom of the conduction band. There are arguments in favor of this view: (i) A kink in the temperature dependence of the ESR linewidth was observed in $YbB_{12}$ at 13–15 K and suggested to be caused by intragap states,[27] coming from exciton-polaronic complexes. (ii) At the same temperature (around 15 K) the activation energy of the charge transport characteristics changes from 10 meV (due to excitations across the hybridization gap) to 2.2 meV;[32,35] the value which basically coincides with the energy position of 2.7 meV of our low-temperature conductivity peak, which can thus be associated with the photon-assisted breaking of excitonic bound states, while the characteristic activation energy of transport properties then corresponds to thermal activation of carriers from excitonic band into conduction band. Thus we suggest that the 22 $cm^{-1}$ peak in the conductivity spectrum of $YbB_{12}$ can correspond to an exciton-polaronic bound state arising from the coupling of free electrons to soft valence fluctuations. Within this picture the "impurity state" can be estimated to have a spatial extension of $r=h^2\epsilon/m^*e^2=5$ Å, close to the lattice spacing of 7.5 Å in $YbB_{12}$;[20] the same value can be also estimated for a small-size Frenkel exciton.[45] More experiments are needed to verify this assumption, as well as to explore the mechanism of how bosonic excitons can be born out of the coherent heavy-fermion condensate.

## V. CONCLUSIONS

In summary, using infrared reflectivity and terahertz transmissivity techniques, optical spectra of $YbB_{12}$ single crystals are studied at frequencies 8 $cm^{-1}$–10 000 $cm^{-1}$ corresponding to quantum energies from 1 meV to 1.2 eV. Typical for free charge carriers a Drude-like response is observed at low energies and characteristics of quasiparticle condensate are correspondingly evaluated. Upon cooling down to $T=70$ K, the condensate relaxation rate strongly decreases indicating a phonon assisted, non-Kondo scattering. Below the coherence temperature of $T^*=70$ K clear indications of a complete coherence among the $4f$-sites are detected: the scattering rate of charge carriers decreases following the Fermi liquid behavior $\gamma \propto T^2$, and their effective mass and the relaxation time increase to values $m^* \approx 34 m_0$ and $\tau=4\times10^{-13}$ s (at $T=20$ K), respectively. At the lowest temperature of 5 K an absorption peak is discovered at 2.7 meV whose origin might be connected to an exciton-polaronic bound state, which has already been detected in a number of intermediate valence semiconductors and can thus be considered as a common characteristic of their ground state.

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