Optical evidence for heavy charge carriers in FeGe

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The optical spectrum of the cubic helimagnetic metal FeGe has been investigated in the frequency range from 0.01 to 3.1 eV for different temperatures from 30 to 296 K. The optical conductivity shows the evolution of a low-energy (0.22 eV) interband transition and the development of a narrow free-carrier response with a strong energy and temperature dependence. The frequency-dependent effective mass and scattering rate derived from the optical data indicate the formation of dressed quasiparticles with a mass renormalization factor of 5. Similar to FeSi the spectral weight in FeGe is not recovered over a broad frequency range, an effect usually attributed to the influence of the on-site Coulomb interaction.

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Cubic FeGe is a good metal at low temperature, which undergoes a transition to helimagnetic order[1] at $T_C=280$ K with the magnetic moment at the iron sites of $1\mu_B$. The helix changes its orientation in a temperature interval $T_2\pm 20$ K and shows pronounced temperature hysteresis[2] between 211 and 245 K. This material crystallizes in the $B20$ structure and the cubic space group $P2_13$ lacking a center of symmetry which is responsible for this long-range order. The isoelectronic compound FeSi has the same crystal structure. It has a large magnetic susceptibility at room temperature, which vanishes as the temperature approaches zero due to a small (70 meV) semiconductor gap at E_F. A continuous series $FeSi_{1-x}Ge_x$ can be formed, where the metal insulator transition[3] occurs for $x\approx 0.25$. Theoretical models, which have been proposed to explain this behavior, invoke disorder,[4] narrow bands, and different ways of incorporating electron correlations.[5–8] The temperature-dependent disappearance of the gap has been explained as a result of a correlation gap using a two-band Hubbard model,[9,10] and excellent agreement was obtained with optical data,[10–12] but it has been shown that vibrational disorder, if sufficiently strong, also closes the gap.[13]

Anisimov *et al.*[14] have predicted a magnetic-field-driven semiconductor to metal transition in $FeSi_{1-x}Ge_x$, and argued that the difference in electronic structure between FeSi and FeGe in essence consists of a rigid relative shift of the majority and minority-spin bands for the latter material. According to this model the optical spectra at low energies are expected to be the superposition of a Drude peak and an interband transition across an energy range corresponding to the forementioned relative shift of the majority and minority bands. Experimentally relatively little is known about the electronic structure of FeGe, for example, no optical data have been published.

Here, we report optical measurements on a cubic FeGe single crystal at different temperatures. The real and imaginary parts of the dielectric function were derived from the reflectivity and ellipsometry measurements. Optical spectra of FeGe reveal the presence of an important interband transition at 0.22 eV and unusual dynamics of the free-carrier charge. In order to clarify the behavior of the optical conductivity the data were compared with local spin-density approximation (LSDA) calculations of the electronic structure.

Cubic FeGe single crystals were grown by chemical-vapor transport method as described in detail in Ref. 15. The single crystals were characterized by transport, magnetic, and thermodynamic measurements. It was found that a first-order phase transition to the helimagnetic state occurs at $T_C=280$ K. Optical properties of FeGe were obtained using spectroscopic ellipsometry at 0.75–3.1 eV and near normal incidence reflectivity spectra were measured in the energy range from 0.01 to 0.85 eV for different temperatures from 30 to 296 K. The complex dielectric function $\epsilon(\omega)=\epsilon_1(\omega)+4\pi i\sigma_1(\omega)/\omega$ on a broad frequency spectral range between 0.01 and 3.1 eV was calculated by combining the two sets of data and using a variational Kramers-Kronig constrained analysis.[16]

Figure 1 shows the reflectivity $R(\omega)$, the optical conductivity $\sigma_1(\omega)$, and the dielectric function $\epsilon_1(\omega)$ of single-crystalline cubic FeGe at a series of temperatures from 30 to 296 K over a broad frequency range. Absolute values of the reflectivity were obtained by calibrating the instrument using a gold layer deposited *in situ* on the sample surface, without breaking the vacuum and without moving the sample. This calibration procedure is designed to fully compensate the frequency dependence of the instrument and the geometry of the sample.

The two distinct sharp excitations in the far-infrared spectrum [230 cm^{-1} (28.5 meV), 290 cm^{-1} (35.9 eV)] are due to optically active phonons.[12] We observe that at low frequency the reflectivity increases with decreasing temperature. In contrast, the range between 500 cm^{-1} (62 meV) and 1500 cm^{-1} (0.18 eV) $R(\omega)$ is strongly suppressed and a dip develops at low temperature. Consequently the far-infrared region of the optical conductivity spectrum displayed in

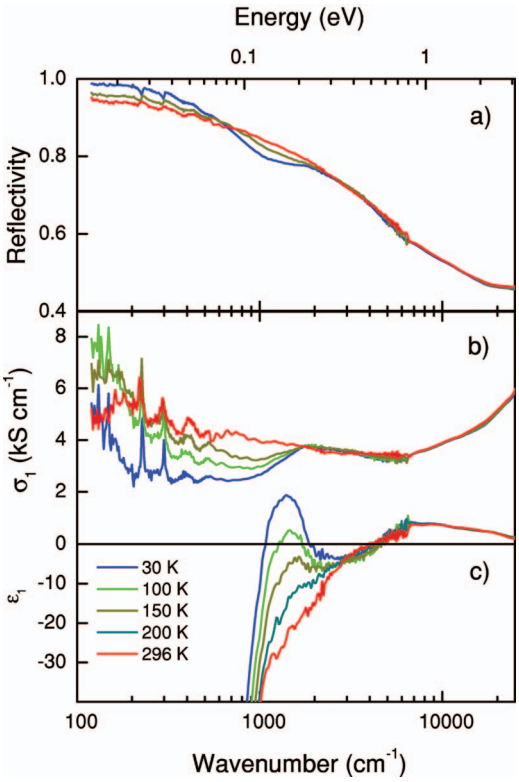

FIG. 1. (Color) The normal-incidence reflectivity $R(\omega)$ (a), optical conductivity $\sigma_1(\omega)$ (b), and real part of the dielectric function $\epsilon_1(\omega)$ (c) of FeGe derived from the ellipsometry and reflectivity measurements at several temperatures ranging from 30 to 296 K on a logarithmic scale.

Fig. 1(b) is strongly temperature dependent. At low temperature $\sigma_1(\omega)$ shows a minimum around 0.1 eV, which vanishes at T_C. Below T_C a peak in the optical conductivity appears at 0.22 eV, which looks like an onset of interband transitions. We observe a narrowing of the free-carrier response while the temperature is lowered to zero. However a large finite conductivity remains below the interband transition at 0.22 eV which appears to be the high-energy tail of the free-carrier response, due to the coupling to bosonic degrees of freedom. In the present case these are most likely spin fluctuations. The narrowing of the free-carrier response signals a strong reduction of the scattering rate, whereas the simultaneous appearance of an interband transition is similar to observations in Kondo lattices such as URu_2Si_2,[17] $CeAl_3$,[18] $CeCoIn_5$, and $CeIrIn_5$.[19]

The dielectric function [see Fig. 1(c)] has a zero crossing at 0.4 eV for all measured temperatures, which we assign to the plasma resonance of the conduction electrons. A second zero crossing occurs below 150 K. This low-frequency crossing is strongly temperature dependent and is shifting toward lower energy as the temperature is decreasing. Such a line shape of $\epsilon_1(\omega)$ resembles the heavy fermion systems, where the low-frequency plasmon is a characteristic feature of the heavy quasiparticles.[19]

We calculated the electronic structure using the linear muffin-tin orbital code[4] with the self-consistent LSDA method, resulting in a ferromagnetic ground state with a magnetic moment of $1\mu_B$ per Fe atom. The density of states, shown in Fig. 2, is consistent with the schematic density of states of Anisimov *et al.* (see Fig. 3 of Ref. 14). The theoretical optical conductivity has been calculated as a sum of all band transitions within 2.7 eV of E_F at 9216 k points of the irreducible Brillouin zone, including the dipole matrix elements. The effect of thermal disorder and zero-point motion on the band structure is introduced as a band broadening, which is assumed to be equal for all bands. The result of disorder is also a smearing of the spectra at higher energies. The parameter for the band broadening is estimated from calculations of disordered supercell calculations for FeSi and FeGe.[13,20]

The comparison between the calculated and measured optical conductivities (see Fig. 3) is not as good as for CoSi and FeSi.[22] Theoretically we find the onset weak interband transition at an energy as low as 80 meV followed by a gradual increase of the optical conductivity to a maximum of 0.4 eV. The experimental data show a minimum at 0.1 eV followed by a peak at 0.22 eV. In addition two peaks are predicted at 1.6 and 2 eV. The experimental high-frequency optical spectrum is less structured compared to the calculated one. The reason could be that at high energy the scattering is very large. We associate the experimental maximum at 0.22 eV with the theoretical peak at 0.4 eV and the higher-energy structures with the theoretical peak predicted at 1.6 eV. It is not uncommon for transition-metal silicides to observe discrepancies between the energy of the measured and local-density approximation calculation peaks of the optical spectrum. The most likely source of these discrepancies are renormalization effects arising from the strong on-site Coulomb interaction of the transition-metal atoms. The resulting vertex corrections are known to result in deep excitonic states, for example, in NiO.[21] While excitons do not form in FeGe the local vertex corrections should still be important and may cause the observed shift of the main absorption features.

We have varied the lattice parameter in the LSDA calculation to see if this would improve the agreement with the experimental data, but the position of the peak at 0.4 eV turned out to be robust.

In order to further analyze the low-frequency behavior we use the extended Drude formalism. This model is only meaningful in the energy region where the optical response is due to mobile carriers and not to the bound ones. The strong temperature dependence of the optical data [Fig. 1(b)] for frequencies below 0.1 eV, naturally assigned to the mobile carriers, suggests that this model can be applied at frequencies lower than 0.1 eV. According to this formalism the scattering rate $1/\tau(\omega)$ and the effective mass $m^*(\omega)/m$ are represented as follows:

$$\frac{m^*(\omega)}{m} + \frac{i}{\omega\tau(\omega)} = \frac{\omega_p^2}{\omega^2[\epsilon_\infty - \epsilon(\omega)]}, \tag{1}$$

where ω_p=2.7 eV is a plasma frequency which acts as scaling factor here, chosen as to give $m^*(\omega)/m$=1 at 296 K and $\omega/2\pi c$=1000 cm^{-1} (0.12 eV). Furthermore ϵ_∞=264 is the

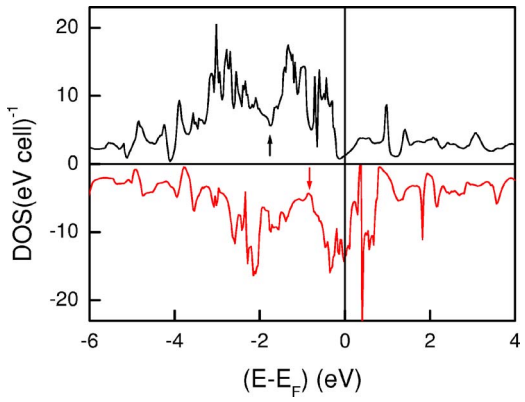

FIG. 2. (Color online) Spin-polarized density of states of FeGe. The energy is relative to E_F.

high-frequency dielectric constant, due to the bound charge polarizability.

Figure 4 displays the spectra of scattering rate and effective mass as a function of frequency for different temperatures. Note that at room temperature both the scattering rate and the effective mass are almost constant. As the temperature approaches the transition temperature, $1/\tau(\omega)$ shows strong frequency and temperature dependences. The value of the scattering rate at high temperature becomes quite large and it seems difficult to describe the metallic state of this material. On the other hand, at low temperature $1/\tau(\omega)$ is much smaller than at room temperature. This behavior of the scattering rate reflects the narrowing of the zero-frequency peak. This suggests that the temperature dependence of the optical response shown in Fig. 1(b) cannot be explained in terms of the simple Drude model. In addition we notice a strong suppression of the scattering rate upon cooling, like in MnSi (Ref. 22) and in the heavy fermion compounds URu_2Si_2,[17] $CeAl_3$,[18] $CeCoIn_5$ and $CeIrIn_5$,[19] suggesting the development of heavy quasiparticles at low temperature. As a result a renormalized Drude absorption due to these heavy quasiparticles is observed in the optical spectra. In relation to this, at low frequency the electrons are dressed by interactions giving them a large effective mass. For $\omega \to 0$ we observe $m^*(\omega)/m \sim 5$. At high frequency the electrons are no longer dressed by the interaction and the effective mass reduces to the band mass as can be seen in Fig. 4(b). In contrast to the case of heavy fermion systems, FeGe has no $4f$ electrons but the large mass renormalization factor appears to be of the same order of magnitude as that observed in $CeCoIn_5$ and $CeIrIn_5$.[19] In this context we speculate that FeGe can be considered as a $3d$ heavy fermion system.

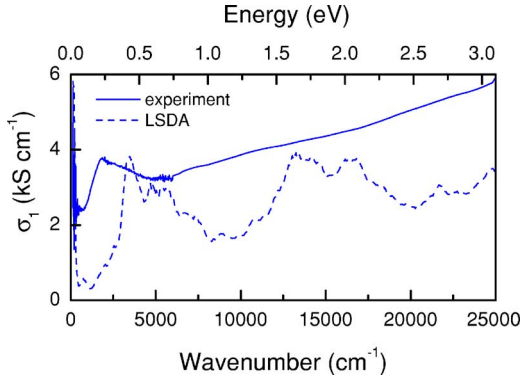

FIG. 3. (Color online) The optical conductivity spectra of the helimagnetic state of FeGe. The solid and dashed lines denote the experimental and calculated optical spectra, respectively.

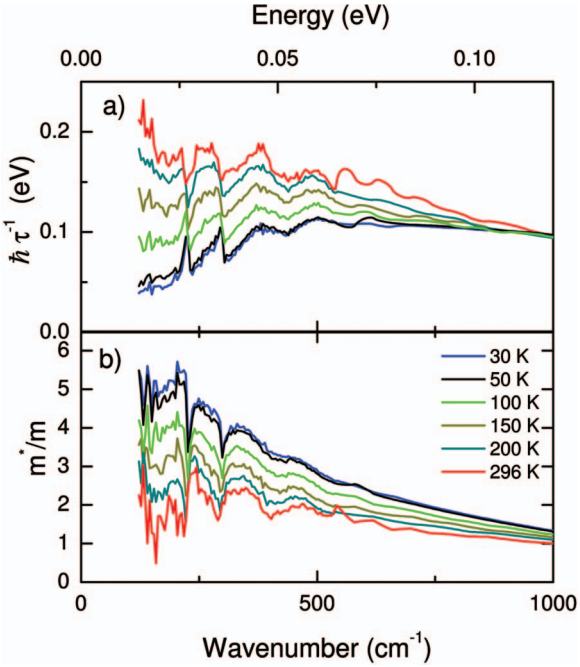

FIG. 4. (Color) Extended Drude analysis of the optical conductivity of FeGe. The details of $1/\tau(\omega)$ above 62 meV are due to the proximity of the onset of interband transitions at 0.1 eV. (ω_p =2.7 eV and ϵ_∞=264 as described in the text).

The half-metallic ferromagnet chromium dioxide has an optical conductivity[23] very similar to that of FeGe. A suppression of $1/\tau(\omega)$ was observed below T_C, which was stronger than in FeGe [Fig. 4(a)]. This is a natural consequence of the half-metallic ferromagnetism: spin-flip scattering, which is the dominant scattering mechanism in ferromagnets, is completely suppressed for frequencies smaller than the gap separating the minority bands from the Fermi level. Note that in *disordered* Co-doped FeSi, which is also a half-metallic

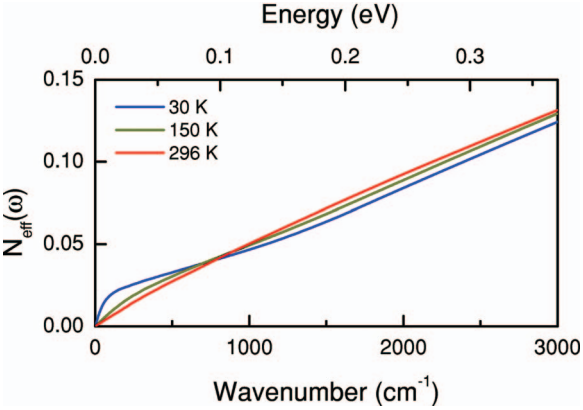

FIG. 5. (Color) The integrated spectral weight of FeGe for a number of representative temperatures.

ferromagnet, the opposite behavior is observed, in that the scattering rate *in*creases in the magnetically ordered state.[24–26]

In FeSi the spectral weight removed at frequencies below 70 meV due to the opening of a gap at low temperatures was observed not to be recovered for energies up to at least 6 eV.[11,26] Since this implies the coupling of the conduction electrons to high-energy scale excitations, possibly on the scale of on-site Coulomb interaction, it may be an indication that the material has features in common with a Kondo lattice, and the insulating gap of FeSi is due to strong local electron correlation effects presumably in the $3d$ shell of the iron atoms. In view of the structural and chemical similarities between FeSi and FeGe one might suspect electron correlation effects to be equally important in the latter material. While FeGe has no insulating gap at low temperatures, the optical spectra depend strongly on temperature. In Fig. 5 the function $N_{eff}(\omega)=\frac{2m_eV}{\pi e^2}\int_0^{\omega_c}\sigma_1(\omega)d\omega$ is displayed. For $\omega_c\rightarrow\infty$ this represents the total number of electrons per unit of FeGe. In the region below 800 cm^{-1} (99.2 meV) we observe at low temperatures that $N_{eff}(\omega)$ increases more sharply as a function of frequency than the high-temperature data, which is a consequence of the fact that $1/\tau$ is smaller and the Drude peak narrower at low temperature. However, all curves cross at 99.2 meV, and above 0.18 eV $N_{eff}(\omega,296\ \mathrm{K})$ exceeds $N_{eff}(\omega,30\ \mathrm{K})$ by a constant amount of $\simeq 0.005$ per FeGe formula unit. In other words, just like in FeSi, cooling down the sample results in a loss of spectral weight in the frequency range 0.1 eV, which is not recovered over a broad frequency range. Due to the smallness of the sample the data noise does not permit a quantitative analysis of the spectral weight transfer for energies larger than 0.5 eV. However, the temperature dependence of $\epsilon_1(\omega,T)$ for $\hbar\omega\approx 1$ eV observed directly with ellipsometry confirms quantitatively the trends seen in Fig. 5. This suggests that in FeGe, just as in FeSi, spectral weight is redistributed over an energy range of order 1 eV or higher when the temperature is varied. This behavior may have its origin in the temperature dependence of the electron correlations resulting from the Hund's rule interaction on the Fe atoms,[10,11] in a change of character of the bands near E_F due to thermal disorder,[4] or in a combination of these two.

In summary, we have reported the optical properties of single-crystalline cubic FeGe, which undergoes a helimagnetic transition at 280 K. At the temperature where magnetic order occurs, a distinct and narrow free-carrier response develops, with a frequency-dependent scattering rate and a moderate mass enhancement in the zero-frequency limit. Similar behavior as in FeSi is observed in FeGe, where the low-energy (Drude) spectral weight appears to be transferred to higher energies, on an energy scale of at least 1 eV, when the material is cooled down. This, together with the observed frequency-dependent mass enhancement, indicates the important role of electron correlations in these materials.

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