d–f exchange resonance and the "*j* polaron" in metallic gadolinium

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The temperature dependences of dynamic conductivity spectra of single-crystalline gadolinium in the region of the *d–f* exchange resonance are investigated experimentally and theoretically. Agreement with experiment is achieved by introducing into the theory quasilocalized electronic states of the *d* type with individual quantization axes at each atom: *j* polarons.

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In the currently generally accepted model of the electronic structure of rare-earth metals (REM), collectivized (c) electrons have with arbitrary temperature a definite z projection of the spin in the coordinate system of the crystal. In ferromagnetic REM, exchange interaction with localized $4f$ electrons leads to splitting of the subbbands of c electrons with opposite spins (Vonsovskiĭ-Zener polarization[1]). In this case, there can be a resonance absorption of light at the frequency

$$\omega_{cf} \sim I_{cf} SM , \tag{1}$$

where I_{cf} is the exchange parameters, $S = 7/2$ is the spin of the $4f$ shell of the RE ion, and M is the magnetic-order parameter. The intensity of the absorption is determined

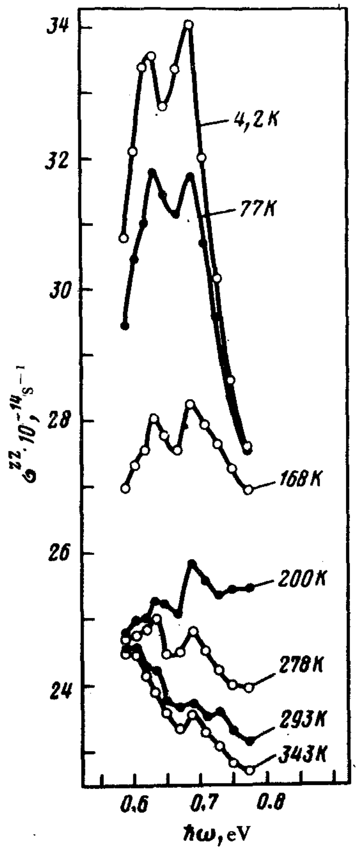

FIG. 1. Temperature family of spectral dependences of the component σ^{zz} of the dynamic-conductivity tensor of gadolinium near the center of the multiplet band of the *d-f* exchange resonance.

by the sign of the dispersion of the *c* electrons and does not depend on the orientation of the easy magnetization axes.

We have obtained, however, experimental data that refutes the universality of the picture with respect to REM. The temperature dependences of the components $\sigma^{zz}(\omega)$ and $\sigma^{yy}(\omega)$ of the dynamic conductivity tensor, investigated with the help of an optical ellipsometrical technique, using a single-crystalline specimen of Gd with high-quality reflecting surface, established that in the magnetically ordered state, the dependences $\sigma(\omega)$ contain "stationary" magnetic multiplet bands in the regions $0.55 < \hbar\omega < 0.8$ eV and $0.5 < \hbar\omega < 0.9$ eV, respectively. For $\sigma^{zz}(\omega)$ and $\sigma^{yy}(\omega)$ (the Z axis corresponds to the direction ⟨001⟩, while the Y axis corresponds to the direction ⟨1010⟩ in the *hcp* lattice of the crystals). The temperature family of the dependences $\sigma^{zz}(\omega)$ in the energy range near the center of the magnetic absorption band is shown in Fig. 1. Figure 2 shows the temperature dependences of $\sigma^{zz}_{0.69}$ and $\sigma^{yy}_{0.71}$ at points corresponding to one of the peaks (for definiteness, the largest one) of these multiplets and the temperature dependence of the angle φ between the direction of the spontaneous magnetic moment

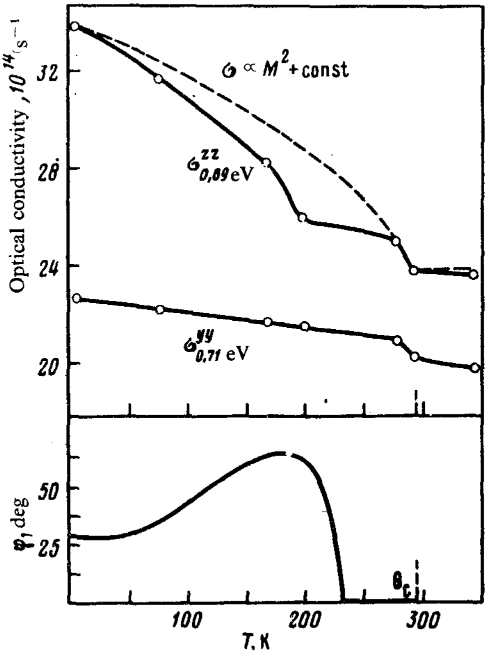

FIG. 2. Temperature dependences of the resonance conductivities $\sigma^{zz}_{0.69}$ and $\sigma^{yy}_{0.71}$ as well as the angle φ between the direction of the spontaneous magnetic moment and the C axis in bulk gadolinium crystal.

and the C axis of the crystal from Ref. 2. As is evident from the figures, magnetic optical absorption bands with centers at frequencies close to the frequency

$$\omega_{df} = \hbar^{-1} 2 I_{df} S = \hbar^{-1} 0.7 \text{ eV} \tag{2}$$

of d–f exchange resonance in the free Gd atom $I_{df} = 0.1$ eV is the $5d$-$4f$ exchange integral in the REM), appear below the Curie point θ_C and grow together with the spontaneous magnetic moment, without a frequency displacement, as would be expected based on relation (1), which is established very accurately according to the position of the peaks in the fine structure of the bands. Fitting simple power-law dependences for $\sigma(M)$ at $\omega = \omega_{df}$ showed that the dependence $\sigma(M) - \sigma(0) \propto M^2$ describes the experimental data best (dashed line in Fig. 2). The deviation from this dependence observed for σ^{zz} correlates with the change in the angle φ. On the whole, the dependences $\sigma^{zz}(\omega_{df})$ and $\sigma^{yy}(\omega_{df})$ are satisfactorily described by the simple equations (in units 10^{14} s^{-1})

$$\sigma^{zz} = (10.59 \pm 3.40) M^2 \cos^2\varphi + (23.53 \pm 1.18), \tag{3a}$$

$$\sigma^{yy} = (2.13 \pm 0.03) M^2 + (20.28 \pm 0.01). \tag{3b}$$

The high intensity of the bands and the polarization of the light reflected from the specimen indicate the electric dipole nature of the optical absorption at the frequency ω_{df}. For this reason, the strong dependence of $\sigma(\omega_{df})$ on φ of the type (3a) is just as puzzling as the satisfaction of relation (2) in the crystal instead of (1).

The dependences (2) and (3) arise in a natural manner in a model of quasibound $5d$ electrons in REM with an individual quantization axis on each atom. This model can be based on the following considerations. We shall distinguish between the intraatomic("optical") oscillations of the spin and orbital moments, due to which the configuration of the system $4f$ and $5d$ electrons within the atom changes, and the interatomic ("thermal") oscillations, due to which the magnetic moment of the pairs of atoms changes. The magnetic moment, however, remains the same in each atom. The frequency of the former is, correspondingly, $\hbar\omega_{df} = 2I_{df}S = 0.7$ eV and $\hbar\omega_{SL} = \zeta s(2L+1) = 0.325$ eV and the frequencies of the latter are $\hbar\omega_q \lesssim 2ZIS = 1.2\times 10^{-2}$ eV. Here $\zeta = 0.13$ eV is the spin-orbit Gd coupling constant, $s = 1/2$ and $L = 2$ are, respectively, the spin and the orbital angular momentum of the $5d$ electron, $I = 1.43\times 10^{-4}$ eV is the interatomic exchange integral in REM, and $Z = 12$ is the number of nearest neighbors. Since $\omega_{df} \sim \omega_{SL} \gg \omega_q$, the optical oscillations will be fine-tuned under considerably slower thermal oscillations of the total angular momentum J of the $4f$ shell, giving the orientation of the local quantization axis of both the spins and of the orbital states of $5d$ electrons. Thus we obtain states analogous to Nagaev's spin-polaron states in magnetic semiconductors,[4] but in this case the corresponding quasiparticle should be called a "j polaron." Using the terminology in Ref. 4, we can say that in such a model of REM the motion of a conduction electron along the crystal is equivalent to the motion of an "irregular" total angular momentum j of the $5d$ electron along the lattice of "regular" angular momenta J of $4f$ shells. Of course, small values of the hopping integral of $5d$ electrons, which are realized effectively in REM, in which there are extensive flat sections of bands both above and below the Fermi level, are favorable for the existence of the j polaron.

In the j-polaron model at frequency ω_{df}, the vector $\mathbf{D}$ of the effective dipole moment of the crystal, which represents the average of the local dipole moments of $5d\uparrow$–$5d\downarrow$ transitions and which is coupled in magnitude and direction to the magnetization M, interacts with an external electric field in the ferromagnetic case. This immediately leads to the dependence $\sigma \propto |\mathbf{DE}|^2 = f(\alpha)M^2\cos^2\alpha$, where $\alpha = \widehat{\mathbf{ME}}$, and $f(\alpha)$ is determined by the mechanism for removing the restriction on the $5d\uparrow$–$5d\downarrow$ transition. Calculating $\sigma(\omega)$, we simplify the problem: we removed one $5d$ electron and one channel of the electric dipole; $5d\uparrow$–$5d\downarrow$ transition, the restriction on which was removed due to the spin-orbit coupling and the odd component of the crystal field. This simplification simplified the calculation, but led to a rough result, especially for σ^{yy}. The computed values of σ^{zz} and σ^{yy} (a paper containing the details of the calculation is being prepared for publication) satisfy condition (2), exhibit a quadratic dependence on M, and give the correct relation between the numerical coefficients in front of M^2:

$$\sigma^{zz}(\omega_{df}) \approx 13.5AM^2\cos^2\varphi, \qquad \sigma^{yy}(\omega_{df}) \approx 2.9AM^2\sin^2 2\varphi,$$

where A is a constant. As we can see, σ^{zz} corresponds to (3a) and σ^{yy} contains the "extra" dependence on φ. This dependence can be weakened by including the dipole mechanisms for removing the restriction on $5d\uparrow$–$5d\downarrow$ transition, for example, by taking into account the dynamic odd component of the crystal field.

As follows from these results, the general features of the j-polaron model introduced in this work agree with experiment and, for this reason, can form the basis for searching for an adequate picture of the electronic structure of REM and other transition metals.

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