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Resonant mode in rare-earth based strongly correlated semiconductors

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Abstract

An extensive study of the spin dynamics in the strongly correlated semiconductors YbB_{12} and SmB_6, as well as in the series of related electron-doped systems, has been performed by means of inelastic neutron scattering spectroscopy. It was found that rare-earth compounds with a valence instability may develop an exciton-like in-gap excitation, generally similar to the so-called *resonant mode* initially discovered in HTSC. The results of this work, along with the literature data for a number of high-T_c-, heavy-fermion- and pnictide superconductors and some magnetic compounds point to such type of resonant excitations being characteristic for systems with gap-like dynamical response in the presence of *competing interactions*. The correlation between the temperature evolutions of the resonant mode and of the gap in the excitation spectrum is discussed.

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Keywords: inelastic magnetic neutron scattering; resonant mode; strongly correlated semiconductor

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1. Introduction

Resonant magnetic excitations in the superconducting state have been discovered [1] and studied in detail [2,3] in the cuprate high-T_c superconductors (HTSC). Then a similar resonant mode (RM) has also been reported for a number of heavy-fermion and iron-pnictide superconductors, such as UPd_2Al_3 [4], $CeCoIn_5$ [5], $CeCu_2Si_2$ [6], $BaFe_2As_2$-based compounds [7,8]. The characteristic RM features are its strong localization in both energy and momentum space and its suppression on increasing the temperature above T_c. The RM is often considered to be hallmark of unconventional superconductivity. However, excitations of this type seem to be more general, and apparently inherent to the systems with gap-like dynamical response and a *competition of different interactions* (for instance, hybridization of *f*- and *d*-electron states, crystalline electric field and inter-site exchange interaction). Recently, RM-like excitations have been observed below the ordering temperature in magnetic $CeRu_2Al_{10}$ [9] and CeB_6 [10]. Moreover, the possible existence of similar excitations has been discussed for semimetals like graphite [11,12].

Magnetic excitations with characteristic properties of RM can be identified in the dynamical magnetic response of the Kondo insulator YbB_{12} [13] and the intermediate-valence compound SmB_6 [14]. In this paper, we review the results of previous inelastic neutron scattering studies of the spin dynamics in YbB_{12} [13,15-18] and SmB_6 [14,19,20] along with a new results, with a special focus on the RM behavior. A crucial piece of information on YbB_{12} has been obtained using polarized neutrons [17]: strong phonon peaks appeared just in the energy range of RM, but finally could be reliably separated by means of the linear polarization analysis.

2. Resonant mode in the Kondo insulator YbB_{12}

Fig.1a shows the magnetic excitation spectrum of YbB_{12} at 5K obtained on single crystal sample [17] using neutron polarization analysis. The characteristic features of this spectrum are the spin gap below 10 meV and the fine structure consisting of three peaks at 15 meV (M1), 20 meV (M2) and 40 meV (M3). In contrast to M2 and M3, M1 has a resolution-limited energy width [13] and is strongly localized in reciprocal space (see Fig.1b) around **q** = (0.5, 0.5, 0.5), which corresponds to the Brillouin zone boundary along the (ξ, ξ, ξ) direction (*L*-point). The overall *Q*-dependence of M1 intensity follows that of the Yb^{3+} magnetic dipole form factor [13,17]. The energy dispersion of M1 is relatively weak but nevertheless demonstrates well-defined minimum at the *L*-point [13,17].

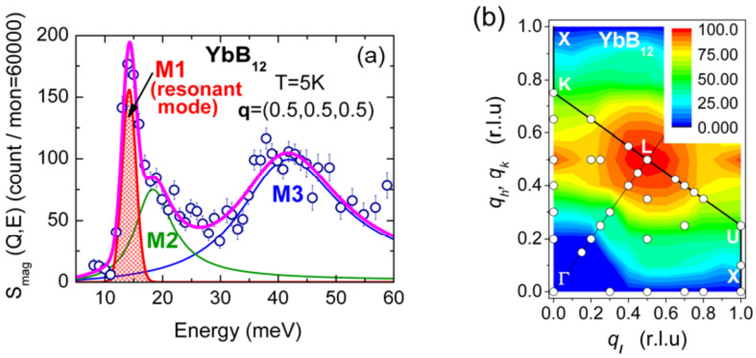

Fig. 1. (a) Magnetic excitation spectrum of YbB_{12} [17] measured on a single crystal at T = 5 K, **q** = (0.5, 0.5, 0.5) (Brillouin zone boundary along the (ξ, ξ, ξ) direction). Symbols: experiment, lines: fit with three spectral component; hatched area: resonant mode. (b) Intensity map (in arbitrary units) at T = 5 K for the 15 meV peak (M1) over one quadrant of the Brillouin zone (adapted from [13,17]); circles denote **Q** vectors at which energy spectra have been measured.

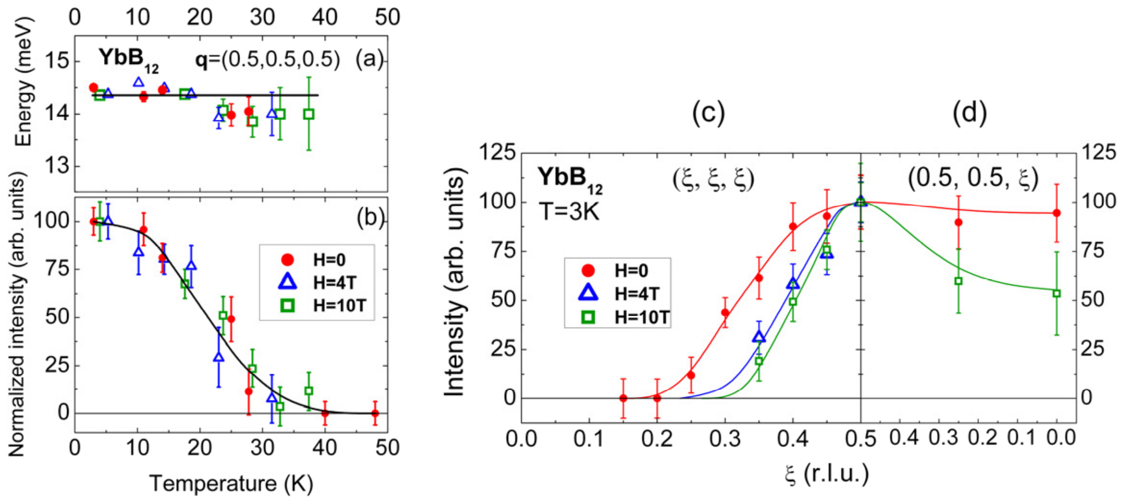

Fig. 2. Temperature and momentum dependence of M1 parameters for different values of applied magnetic fields ($H = 0$, $H = 4$ T, $H = 10$ T). (a,b): evolution of M1 energy (a) and integrated intensity (b) at **q** = (0.5, 0.5, 0.5). (c,d): Q-dependence of M1 integrated intensity along (ξ, ξ, ξ) (c) and (0.5, 0.5, ξ) (d) directions. Lines – guides to the eye. All dependencies in frames (b,c,d) for $H = 4$ T and $H = 10$ T are normalized to the maximum intensity in zero field (the value for **q** = (0.5, 0.5, 0.5) at $T = 3$ K).

On increasing temperature, the magnetic excitation spectrum starts to transform [13,17]: the spin gap is gradually filled by the broad ($\Gamma \approx 15$ meV) quasielastic signal, whereas M1, M2 and M3 are concurrently suppressed. The transformation to the high-temperature spin-fluctuation regime (with residual crystal-field effects) is completed near $T \sim 100$ K. However, M1 becomes undetectable already at $T \sim 50$ K (see Fig. 2b). The energy of M1 is almost temperature independent within its whole range of existence, with a slight decrease above $T = 20$ K (see Fig. 2a).

In the experiment on the electron-doped system $Yb_{1-x}Zr_xB_{12}$ [18], it was shown that M1 is closely connected with the existence of the spin gap. Filling of the spin gap due to the substitution of Yb by Zr results in the suppression of M1. On the other hand, M1 seems to be less robust than the spin-gap itself. In particular, the isoelectronic substitution of Yb by Lu does not influence to the spin-gap [15,16]. Nevertheless, at high concentration of Lu (above 75%) M1 is suppressed (essential broadening of M1 takes place already at lower Lu-concentrations). In this sense, the effect of doping is rather similar to that of temperature.

Approximately in the same temperature range where the spin-gap regime transforms to the spin-fluctuating one, the transition from a semiconducting to a metallic state occurs. The filling of the gap in the electron density of states was clearly observed in the temperature evolution of the dynamical conductivity [21]. Taking into account the above-mentioned properties of M1, this excitation can be ascribed to a RM, which characterizes the low-temperature semiconducting non-magnetic state in YbB_{12}. In analogy with one of the interpretations for the RM in the cuprate high-T_c superconductors [22], this excitation is proposed to be a spin exciton which is split off from the excitation continuum due to the dynamical antiferromagnetic (AFM) exchange interaction between 4f electrons [23,24]. From this point of view, M1 should be considered as the *in-gap* excitation, in good agreement with the very low intrinsic energy width indicative of a long relaxation time. The broader peak M2 is most likely associated with transitions across the gap, and the spin gap value can be estimated to be ~ 20 meV [17].

In order to check the antiferromagnetic origin of the RM in YbB_{12}, we have studied the influence of an applied magnetic field on its parameters. The measurements have been performed at the ILL on the triple-axis spectrometers IN22 and IN8 in the temperature range 3–50 K, with magnetic fields up to 10 T. The spectra with and without external magnetic field have been recorded in the same conditions and analyzed by the same procedure.

According to literature data [25], a magnetic field of 10 T reduces the insulating gap only slightly. Therefore, changes in the excitation spectrum under the application of a magnetic field are expected to reflect the evolution of exchange interactions. One straightforward effect would be a Zeeman splitting of the resonance peak (e.g., if the RM corresponds to a singlet-triplet transition). However, we have observed no clear splitting of the RM in YbB_{12}, though some energy broadening could be indicative of a minor effect. Careful study of this broadening will be the subject of future experiments. For now, the main result of the application of the magnetic field is a reduction of the RM intensity. At low temperature this reduction is about 15% at the *L*-point for $H = 4$ T, and 30% for $H = 10$ T. To trace the influence of the magnetic field on the temperature and Q dependencies of the RM, we have normalized all dependencies to the maximum intensity in zero field (the value for $\mathbf{q} = (0.5, 0.5, 0.5)$ at $T = 3$ K).

The results of the data treatment are shown in Fig. 2. Surprisingly, the magnetic field does not influence the temperature dependence of the RM energy (Fig. 2a) nor of the intensity (Fig. 2b). On the other hand, we have observed a significant enhancement of the RM localization in reciprocal space (Figs. 2c, 2d). This is clearly seen along the (ξ, ξ, ξ) direction and especially along $(0.5, 0.5, \xi)$, where the RM intensity was almost constant in zero field. This effect confirms the magnetic origin of the RM in YbB_{12} and can be understood in terms of the correlation length of the dynamical AFM interaction underlying the spin exciton. If we assume the presence of regions with different correlation lengths, those with the shortest correlation lengths should be the least robust against the applied magnetic field. Thus, the narrowing of the peak in Q space could denote the suppression of the AFM correlations having the shortest correlation lengths. This suggestion is also consistent with the suppression of the total RM intensity mentioned above.

3. Resonant mode in SmB_6: system with essentially intermediate valence

A RM-like excitation with quite different properties has been found in SmB_6 [14]. Similar to YbB_{12}, this compound undergoes a transition from semiconducting to metallic state at $T^* \sim 50$ K, with an electronic gap which is filled rather than closed as T increases [26]. The specific feature of SmB_6 is the essentially intermediate valence of the Sm ions ($v \approx 2.55$ [27]).

The RM in SmB_6 is a resolution-limited peak with strong localization near the Brillouin zone boundary at $\mathbf{q} = (0.5, 0.5, 0.5)$ (see Fig. 3a). However, in contrast to YbB_{12}, the only $\mathbf{Q}$ vector at which the RM was observed in SmB_6 is $\mathbf{Q} = (0.5, 0.5, 0.5)$, and the peak is not reproduced at the multiple Q vectors. The steep Q dependence of the RM intensity in SmB_6 is in agreement with a delocalization of the *f*-electron shell due to the intermediate valence state. The role of the Sm valence is illustrated in Fig. 3b for the series of SmB_6-based samples with partial substitution of Sm by La, where the Sm valence is shifted towards 2+ on increasing the La concentration. One can see that the Q dependence becomes less steep as the La concentration increases (Sm valence decreases), but it gives no indication of having a maximum at the next Brillouin zone boundary or minimum at the zone centre expected for an AFM spin-exciton of the type discussed above. Thus, despite some similarities of the RM in SmB_6 and YbB_{12}, their origins seem to be quite different. Indeed, the behavior of the RM in SmB_6 could be described quantitatively using the excitonic model of the intermediate valence proposed by Kikoin and Mishchenko [28]. In this model the RM is interpreted as analogue of the ${}^7F_0 \rightarrow {}^7F_1$ spin-orbit transition of the parent Sm^{2+} configuration that renormalized due to formation of the intermediate-valence state.

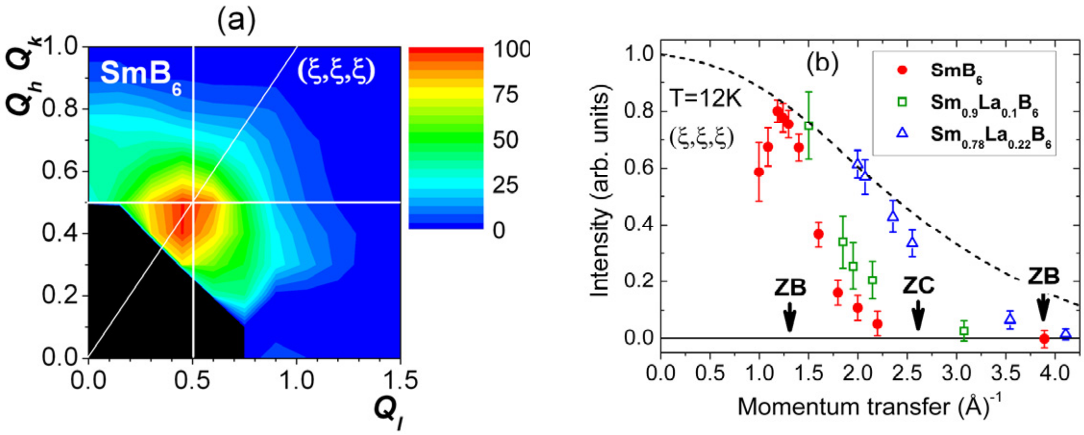

Fig. 3. Resonant mode in SmB_6. (a) Intensity map (in arbitrary units) at $T = 12$ K for the resonant mode over one quadrant of the Brillouin zone (adapted from [20]). (b) Q dependence of the resonant mode intensity in SmB_6, $Sm_{0.9}La_{0.1}B_6$, and $Sm_{0.78}La_{0.22}B_6$ along (ξ, ξ, ξ) direction at $T = 12$ K (combined data from [14,19,20]). Dashed line: magnetic dipole form factor for the $^7F_0 \rightarrow {}^7F_1$ spin-orbit transition of Sm^{2+}. Vertical arrows at the bottom of the frame indicate the values of momentum transfer corresponding to the Brillouin zone boundaries (ZB) and Brillouin zone centres (ZC).

4. Discussion and concluding remarks

In this section we briefly compare the behavior of the “classical” RM in unconventional superconductors, RM that occurring in some magnetic compounds and described above RM in strongly correlated semiconductors. The most obvious feature of those different types of RM is a gradual suppression of their intensity upon heating to some transition, or crossover, temperature, which is somehow related to the opening of an electronic gap. More intriguing is the temperature evolution of the RM *energy*. In the majority of these systems [5,6,8-10], the suppression of the gapped state with increasing temperature causes a gradual reduction of the RM energy, like the order parameter at the phase transition. The exception is the RM in cuprate superconductors, whose energy remains almost constant within the whole temperature range of existence [29]. In this sense, the RM in YbB_{12} and SmB_6 is similar to RM in those HTSC that is especially striking in view of the entirely different origins of the RM and of the gap itself. Presumably, such a behavior of the RM reflects the mechanism by which the gap develops. In the systems where the electronic or the spin gap opens gradually, the RM energy simply follows the energy of the gap edge. However, if the gap develops abruptly, like in cuprate HTSC [2,3], the RM energy also remains nearly constant. Thus, a detailed study of the RM could give a way to trace the development of the spin gap and even the *charge* gap by means of inelastic neutron scattering. Close connection between the resonant *magnetic* excitation and the *charge* excitation spectrum is well known for superconductors [3] where a feedback from the RM manifests itself as the so-called *“peak-dip-hump”* feature in the angle-resolved photoemission and tunneling spectra. A quite similar effect has been reported for SmB_6 [30,31], where the RM is directly related to the charge subsystem via the formation of an excitonic intermediate-valence state. Some indication of the existence of a peak in the photoemission spectra of YbB_{12} at nearly the same energy as the RM has also been reported [31,32].

In conclusion, it is worth noting that the RMs found in YbB_{12} and SmB_6 are not isolated cases among Kondo-insulator-like systems. Basing on our extensive study of YbB_{12} and SmB_6, we have identified damped

RMs in the dynamical magnetic response of the metallic spin-gap systems $Sm_{1-x}Y_xS$ [33,34] and $EuCu_2(Si_xGe_{1-x})_2$ [35]. Excitations discovered long ago in the semimetallic heavy-fermion compound CeNiSn [36] with an anisotropic hybridization gap might also be of RM type, though this possibility has not yet received sufficient consideration. Even in the archetype Kondo insulator $Ce_3Bi_4Pt_3$, where no RM has been observed so far in powder experiments [37], one cannot exclude that such a feature may have been smeared out by *Q*-space averaging.

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