Proc. Int. Symposium on Advances in Neutron Scattering Research, Tokai, 2000
J. Phys. Soc. Jpn. **70** (2001) Suppl. A pp. 224–226

Magnetic Excitations in the Zn-Mg-RE Icosahedral Quasicrystals

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Neutron inelastic scattering experiments were performed on the Zn-Mg-Tb icosahedral quasicrystals. It was found that the observed spectra consist of quasielatic scattering and a broad inelastic-scattering peak centered at $E \sim 2$ meV. The inelastic part is almost independent of Q, indicating a possibly-localized excitation in the icosahedral quasicrystal. Whereas for the quasielastic part, the scattering intensity, $S(Q,E)$, at the lowest temperature decreases but remains finite for $E \to 0$ meV. This suggests vanishing $\chi''(E)$, the imaginary part of the dynamical susceptibility, for $E \to 0$ meV even below the macroscopic spin-glass-transition temperature.

KEYWORDS: neutron scattering, Zn-Mg-RE, icosahedral quasicrystal, magnetic excitation

§1. Introduction

Quasicrystals have no periodicity in spatial atomic arrangement, instead have so-called *quasiperiodicity*, which is defined as a projection of a higher dimensional periodicity into the real three-dimensional space.[1)] It is, thus, an intriguing issue to elucidate behavior of spin systems in the novel quasiperiodic structures. The Zn-Mg-RE (RE: rare-earth) icosahedral quasicrystals[2)] are the first example of quasicrystals with well-defined magnetic moments originating from $4f$ electrons of the RE atoms, and thus provide a unique opportunity to study the behavior. A number of studies were performed on the Zn-Mg-RE quasicrystals to date. In short, the magnetic susceptibility indicates a spin-glass-like freezing at low temperatures.[3)] On the other hand, the neutron diffraction study detected significantly-developed static short-range-spin correlations at the lowest temperature.[4,5)] It is now known that the spin correlations can be regarded as a projection of virtual short-range spin correlations defined in the six-dimensional space,[6)] where the icosahedral quasicrystal is mapped into a periodic crystal. Therefore, the correlations are believed to be intrinsic to the quasiperiodic structure.

Since the static spin correlations are quite unconventional, dynamics of the spins may also essentially differs from those observed in the ordinary spin glasses. In addition, information on the dynamics may possibly give some insight into the origin of the spin-freezing in the icosahedral quasicrystals. Thus, detailed neutron-inelastic-scattering study has been highly desired. To date, only two studies along this line were performed and gave somewhat contradicting results; the powder neutron scattering suggests an inelastic excitation peak at $E \sim 0.35$ meV,[7)] whereas the single quasicrystal study infers only quasielastic scattering,[8)] both in the Zn-Mg-Ho quasicrystal. Details of the magnetic response have not been explored at all. In the present study, we have performed neutron inelastic scattering experiments using powder samples of the Zn-Mg-Tb quasicrystal. The Zn-Mg-Tb quasicrystal has a considerably larger characteristic energy scale than that of the Zn-Mg-Ho quasicrystal, inferred by their freezing temperatures: $T_f \sim 5.6$ K for Zn-Mg-Tb; $T_f \sim 1.95$ K for Zn-Mg-Ho. Therefore, it is expected that the quasielastic and inelastic scattering can be distinguished much clearly. Details of this study will be published elsewhere.[9)]

§2. Experimental

Polycrystalline samples of the $Zn_{57}Mg_{34}Tb_9$ quasicrystals were prepared by melting constituent elements in an induction furnace using Al_2O_3 crucible under an inert Ar gas atmosphere. Purities of the starting elements were 99.9999 %, 99.99 % and 99.9 % for Zn, Mg and Tb, respectively. The polycrystalline alloys were annealed at $T = 773$ K for 100 h to obtain single-icosahedral-phased samples. The neutron scattering experiments were performed at the LAM-40 spectrometer installed at the KENS spallation source, KEK, Japan. Pyrolytic graphite (002) reflections were used for fixing the final energy to $E_f = 4.59$ meV. Energy resolution at the elastic position was estimated as $\Delta E = 320\ \mu$eV (FWHM) using sample-shaped vanadium. The samples were mounted in the standard orange cryostat with the lowest working temperature being about 1.4 K. Background was estimated by a proper combination of empty-cell and sample-shaped-Cd runs, and absorption was corrected by estimating it using numerical integration.

§3. Results and Discussion

Shown in Fig. 1 are the representative spectra observed at $T = 1.4$, 30 and 100 K using the LAM-40 spectrometer. The scattering angle, 2θ, is about 36°, corresponding to $Q \sim 0.93\text{Å}^{-1}$ at the elastic position. The result for the sample-shaped vanadium standard is also shown as an estimate of the instrumental resolution. At the lowest temperature $T = 1.4$ K, the spectrum clearly shows a broad peak centered at $E \simeq 2.5$ meV together with a sharp resolution-limited peak centered at $E = 0$ meV. It can be further seen that the scattering intensity continuously decreases for $E \to 0$ meV, but remains finite at the lowest energy of $E \simeq 0.5$ meV. This remaining intensity at the lowest temperature is preliminarily confirmed by using the LAM-80ET spectrometer down to 50 μeV.[9] Therefore, it is strongly suggested that the finite intensity remains down to $E = 0$ meV.

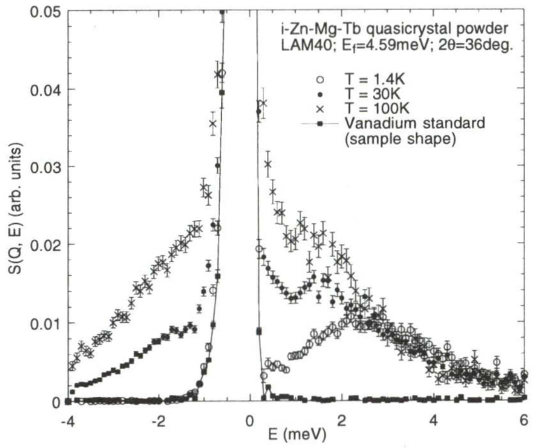

Fig. 1. Neutron scattering spectra observed at $T = 1.4$, 30 and 100 K using the LAM-40 spectrometer. The result for the vanadium standard is also shown.

To gain much information on the origin of the broad peak and on the remaining scattering intensity at lower energies, we, next, study the temperature dependence of the spectra. As also shown in Fig. 1, the scattering intensity for $E < 2.5$ meV drastically increases at higher temperatures. One can also observe concomitant increase of a tail around the elastic peak. On the other hand, the intensity for $E > 2.5$ meV is almost independent of temperature. We note that the neutron scattering intensity, $S(Q, E)$, can be related to the imaginary part of the susceptibility, $\chi''(E)$, or the spectral weight function, $F(E)$, as,

$$S(Q, E) \propto |f(Q)|^2 \frac{1}{1 - \exp(-\beta E)} \chi''(E), \qquad (3.1)$$

$$\propto |f(Q)|^2 \frac{E}{1 - \exp(-\beta E)} \chi_Q F(E), \qquad (3.2)$$

where $f(Q)$ and χ_Q are the magnetic form factor and the isothermal susceptibility, respectively. This relation suggests that the temperature dependence for $E < 2.5$ meV may be attributed to the temperature factor in eq. (3.1) or (3.2). To confirm this point, $\chi_Q F(E)$ is estimated from the data in Fig. 1, and is shown in Fig. 2. For the estimation, we assume that $f(Q)$ is the only Q-dependent term in $S(Q, E)$, *i.e.*, χ_Q is independent of Q, for simplicity. (Note: Q-independence of the inelastic scattering peak will be displayed in the next paragraph.) As can be seen in the figure, shape of the spectral weight shows only slight change for the present temperature range. Whereas, the intensity decreases considerably as temperature is increased, in qualitative accordance with the temperature dependence of the static susceptibility.[3] It is further inferred from the figure that the spectral weight consists of two components: quasielastic scattering centered at $E = 0$ meV and an inelastic peak around $E \sim 2$ meV. It is, in fact, possible to reasonably fit the observed spectral weight using the following Lorentzian-type functions:

$$F(E) = c_0 \frac{\Gamma_0}{\Gamma_0^2 + E^2} + c_1 \left(\frac{\Gamma_1}{\Gamma_1^2 + (E - E_1)^2} + \frac{\Gamma_1}{\Gamma_1^2 + (E + E_1)^2} \right). \qquad (3.3)$$

The results of the fitting are shown in the figure; the obtained parameters are listed in Table I. It may be noteworthy that the inelastic peak energy slightly shifts toward higher energy as temperature is decreased. The width of the quasielastic peak also seems to decrease at lower temperature, however, because the estimated widths are comparable to the instrumental resolution, they are quite ambiguous. Experiments with a higher energy resolution are in progress.

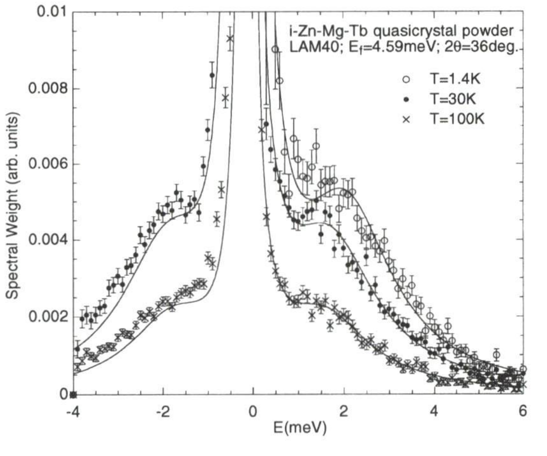

Fig. 2. Spectral weights estimated from the data shown in Fig. 1. Lines represents fit using eq. (3.3).

Table I. List of parameters obtained by fitting the spectral weights.

Temp. (K)	Γ_0 (meV)	Γ_1 (meV)	E_1 (meV)
100	0.25 ± 0.02	1.2 ± 0.1	1.7 ± 0.1
30	0.28 ± 0.07	1.2 ± 0.1	1.8 ± 0.1
1.4	0.17 ± 0.2	1.2 ± 0.1	2.2 ± 0.1

Finally, we check the Q-dependence of the inelastic peak. For this purpose, we have collected several spectra at different 2θ and temperatures using the LAM-40 spectrometer. Then, the scattering intensity was integrated around the inelastic peak positions, *i.e.*, $2 < E < 3$ meV for $T = 1.4$ K, or $1 < E < 2$ meV for $T = 30$ and 100 K, as a measure of the intensity of the inelastic peak. Figure 3 displays the Q-dependence of the E-integrated intensity. In addition, temperature difference of the elastic intensity, $I_{el}(1.4\text{ K}) - I_{el}(100\text{ K})$, is shown in the figure, where I_{el} is obtained by integrating the spectra for $-0.5 < E < 0.5$ meV. The Q-dependence of the elastic intensity reproduces the prior results.[4,5] On the other hand, the inelastic peak shows little Q-dependence, which can be perfectly explained by the Tb^{3+} form factor as shown by the lines in the figure. Hence, it can be concluded that the inelastic scattering peak is independent of Q.

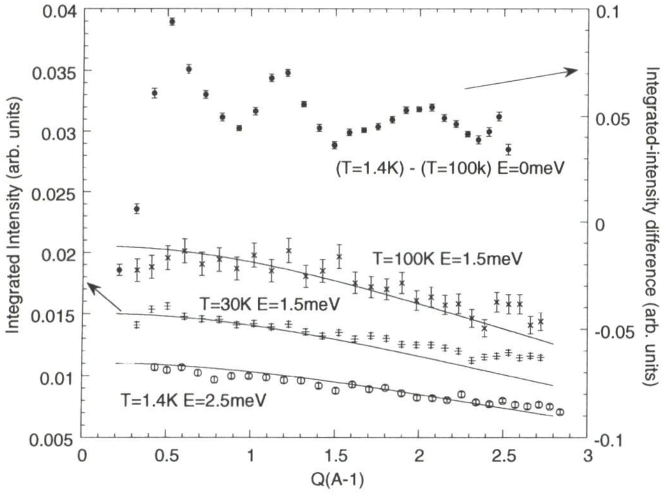

Fig. 3. Q-dependence of the integrated intensities of inelastic scattering at T = 1.4, 30 and 100 K. The temperature difference of the integrated intensity around the elastic position, $I_{\mathrm{el}}(1.4\ \mathrm{K}) - I_{\mathrm{el}}(100\ \mathrm{K})$, is also shown. The lines represent the Tb^{3+} form factor.

As described above, the neutron inelastic scattering observed in the Zn-Mg-Tb icosahedral quasicrystal can be regarded as the superposition of the Lorentzian-shaped inelastic and quasielastic scattering. Here, we will briefly discuss their origin.

As concern the inelastic peak, in view of its Q-independence, a possible origin can be an excitation between (quasi)crystalline electric field (CEF) splitting levels. In general, however, quasicrystals have a variety of local environments differing site to site. Thus, it may be unlikely to have one well-defined excitation levels at about 2 meV as observed. In addition, provided that there are two states at $E = 0$ and 2 meV, then the scattering intensity for the transition $E = 0 \to 2$ meV will decrease by a factor of 0.56 on increasing temperature from 1.4 K to 100 K. In contrast, the intensity at $E > 2.5$ meV shows little change in the Zn-Mg-Tb quasicrystal. Therefore, the CEF origin of the inelastic peak may presumably be discounted, although the above crude estimation cannot perfectly rule out the possibility to explain the observation using much complicated CEF splitting levels. With regard to the quasielastic scattering, similar scattering can be found in the ordinary spin-glasses such as the CuMn system,[10] which is attributed to a wide distribution of relaxation time. There are, however, one important dissimilarity; the spin glasses shows finite $\chi''(E)$ for $E \to 0$ meV, whereas the Zn-Mg-Tb quasicrystal shows decreasing $S(Q, E \to 0)$, which, with the help of eq. (3.1), corresponds to the vanishing $\chi''(E \to 0)$. Thus, both the scattering suffers some difficulty to be related to the well-known mechanisms. Further study is apparently necessary to elucidate the nature of the spin dynamics in the Zn-Mg-Tb quasicrystals.

In summary, the neutron scattering experiments were performed on the Zn-Mg-Tb icosahedral quasicrystals. It was found that the shape of the spectral weight function has little dependence on temperature. The spectral weight consists of two parts: quasielastic scattering centered at $E = 0$ meV and inelastic scattering centered at $E \sim 2$ meV.

Acknowledgement

The authors thank Drs E. Abe and H. Mamiya for valuable discussions.

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