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Abstract

Full Text

Physics

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Paramagnetic Resonance of Cerium in Single Crystals of PbMoO₄

(Presented by Academician E. K. Zavoisky on 10 X 1964)

Several papers have already been devoted to the electron paramagnetic resonance (EPR) of rare earths in crystals of the scheelite type ^(1,2). In the present communication we set forth the results of an investigation of the EPR of the simplest of the rare-earth ions—cerium (Ce³⁺, 4f¹, ²F_{5/2})—introduced into a PbMoO₄ single crystal (scheelite structure). The measurements were carried out on a specimen grown by pulling from the melt and containing nominally 0.3 mol.% Ce³⁺ and Yb³⁺, introduced into the melt in the form of CeO₂ and Yb₂O₃. To compensate the excess charge, a corresponding amount of Na₂MoO₄ was added to the melt.

Despite the fact that the specimen was rather imperfect, the paramagnetic resonance of Ce³⁺ and Yb³⁺ at 4.2°K was clearly observed. The *g*-factors of Ce³⁺ and Yb³⁺ proved to be such that the Yb³⁺ lines did not overlap the Ce³⁺ line and not only did not interfere with the observations, but, on the contrary, made possible an additional check of the orientation of the crystal.

If it is assumed that cerium substitutes for lead, with the symmetry of the nearest environment *S*₄ being preserved, then the ground state of Ce³⁺ ²F_{5/2} should give three Kramers doublets.

We observed one intense line belonging to Ce³⁺. This means that at 4.2°K only one of the doublets is populated. The position of this line is described by the simple spin Hamiltonian (all isotopes of natural cerium have *I* = 0)

$$\mathcal{H} = g_{\parallel}\beta H_{zS}z + g_{\perp}\beta(H_{xS}x + H_{yS}y),$$

where the effective spin *S* = 1/2 and

$$|g_{\parallel}| = 2.684 \pm 0.005, \quad |g_{\perp}| = 1.514 \pm 0.015. \quad (1)$$

The wave functions of the system with *J* = 5/2 in a tetragonal field have the form

$$\Gamma_{t7} : a|\pm 5/2\rangle + b|\mp 3/2\rangle, \quad (2)$$

$$\Gamma_{t6} : |\pm 1/2\rangle.$$

Hence

$$g_{\parallel}(\Gamma_{t6}) = g, \quad g_{\perp}(\Gamma_{t6}) = 3g; \quad (3)$$

$$g_{\parallel}(\Gamma_{t7}) = g(5a^2 - 3b^2), \quad g_{\perp}(\Gamma_{t7}) = 2g\sqrt{5}ab, \quad (4)$$

where $g = 6/7$.

It is clear that the ground state can in no way belong to Γ_{t6} , since in that case there would not even be qualitative agreement with experiment. Indeed, experiment gives $g_{\parallel} \sim 2g_{\perp}$, whereas from (2) it follows that $g_{\parallel} = 1/3 g_{\perp}$.

Thus, the wave function of the ground doublet is transformed according to the irreducible representation Γ_{t7} . The smallest deviation ($\sim 4\%$) from the experimental g -factors is obtained for $|a| = 0.883$:

$$\Gamma_{t7}, \quad a^2 = 0.78; \quad g_{\parallel} = +2.777, \quad |g_{\perp}| = 1.587.$$

The EPR data make it possible to express certain judgments about the quality of the crystals studied and about the nature of their imperfections. A specimen of volume $\sim 170 \text{ mm}^3$ gave an EPR line of distorted shape, indicating the superposition of several curves with nearby maxima. In all, the curve occupied a range of about 150 Oe. When the specimen volume was reduced to $\sim 40 \text{ mm}^3$, the line narrowed ($\sim 50 \text{ Oe}$ for $\mathbf{H} \perp z$ and $\sim 15 \text{ Oe}$ for $\mathbf{H} \parallel z$) and became more symmetrical. It follows from this that the crystal consisted of fairly perfect blocks several cubic millimeters in volume, disoriented relative to one another. The degree of disorientation can be estimated (very roughly) as $0.5\text{--}1.5^\circ$. In turn, each of the blocks also has small imperfections, which are manifested, for example, in the fact that at $\theta = 90^\circ$ the line width depends on φ (θ is the angle between c and \mathbf{H} ; φ is measured in the ab plane from an arbitrary direction in this plane). In this case the change in width is not accompanied by distortion of the line, the appearance of "shoulders," etc. The phenomenon described is apparently connected with mosaicity—lattice disturbances within each block that lead to the occurrence of a large number of weakly disoriented magnetic centers. Estimation of the mosaicity from the EPR data and establishment of its connection with the growth conditions require additional investigations.

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