MAGNETIC PROPERTIES OF SINGLE-CRYSTAL AND POLYCRYSTALLINE PRASEODYMIUM DISELENIDE $PrSe_2$

V. I. CHECHERNIKOV, A. V. PECHENNIKOV, V. I. KALITIN, and E. I. YAREMBASH

Moscow State University

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The magnetic properties of single-crystal and polycrystalline praseodymium diselenide were investigated in the temperature range 77–300°K. It was established that, along the direction parallel to the c-axis of a single crystal, the magnetic susceptibility was higher than at right-angles to this direction. It was also found that the effective atomic magnetic moment P_p of the compound corresponded to the magnetic moment of the triply charged praseodymium ion ($3.62\,\mu_B$) and the paramagnetic Curie point Θ_p had a negative value for the polycrystalline sample and for the single crystal along the direction perpendicular to the c-axis.

1. The magnetic properties of rare-earth metal selenides have been investigated so far using polycrystalline samples only.[1] No experimental data are available on the magnetic properties of single crystals of these compounds. This is primarily because the preparation of single crystals of rare-earth metal selenides is difficult.

Nevertheless, the investigation of the magnetic properties of a rare-earth metal selenide can yield information on the magnetic and electron structure of the compound itself and of the rare-earth metal, in our case praseodymium (the published data on Pr are very contradictory). Moreover, using the data on the magnetic properties, one can determine more exactly the nature of the chemical binding in rare-earth metal selenides.

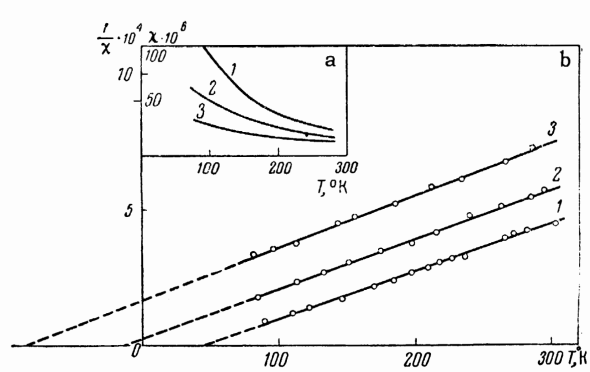
2. The present investigation deals with the temperature dependence of the magnetic susceptibility of single-crystal and polycrystalline praseodymium diselenide at low temperatures (77–300°K).

A polycrystalline sample of $PrSe_2$ was synthesized in an ampoule from the elements praseodymium and selenium, whose purities were 99.4% and 99.99%, respectively. Single crystals of $PrSe_2$ were prepared by the method of chemical transport reactions[2] using iodine as a carrier and employing the diffusion method with a temperature gradient in the reaction space $T_2 \rightarrow T_1$ ($T_2 > T_1$). Polycrystalline Pr_2Se_2 was used as the raw material from which the single crystals were prepared. The single crystals prepared by this method were in the form of truncated tetragonal pyramids, whose dimensions were $0.5 \times 1.5 \times 2$ mm. X-ray structure investigations have shown that $PrSe_2$ belongs to the tetragonal system with the lattice parameters $a = 8.28 \pm 0.007$ Å and $c = 8.44 \pm 0.007$ Å and the ratio $c/a = 1.023$.

A single-crystal sample was placed in a holder in such a way that the magnetic field was directed either parallel or perpendicular to the c-axis. The single crystal was aligned by the Laue method.

The temperature dependence of the magnetic susceptibility was determined by a ponderomotive method, using a pendulum balance.[3] The temperature of the sample was determined by a copper-constantan thermocouple, one of whose junctions was placed next to the sample.

The figure shows the temperature dependence of the specific magnetic susceptibility χ and its reciprocal $1/\chi$ for a single crystal and for a polycrystalline sample. It is evident that the magnetic susceptibility measured along the direction parallel to the c-axis (curve 1) is considerably higher than that along the direction perpendicular to the



Temperature dependence of χ (a) and of $1/\chi$ (b) for a single crystal (curve 1 – H ∥ c, curve 3 – H ⊥ c) and for a polycrystalline sample of $PrSe_2$ (curve 2).

c-axis (curve 3), while the magnetic susceptibility of the polycrystalline sample of $PrSe_2$ is approximately equal to half the sum of the susceptibilities along these two directions in the single crystal. In all three cases, the Curie-Weiss law $\chi = c/(T - \Theta_p)$ is satisfied and the slope of the straight lines $1/\chi = f(T)$ is the same. Calculations show that the effective atomic magnetic moment P_p of the single crystal and of the polycrystalline sample is $3.62\,\mu_B$, which agrees with the theoretical value for the trivalent praseodymium ion in the ground state $^3H_4$.

The most interesting results were obtained in the investigation of the temperature of the paramagnetic Curie point. The experiments showed that the sign of Θ_p depended on the direction of the magnetic field with respect to the crystallographic axes:

$$(\Theta_p)_{\parallel} = 45^\circ\,\mathrm{K}, \quad (\Theta_p)_{\perp} = -90^\circ\,\mathrm{K}, \quad (\Theta_p)_{\mathrm{polycryst}} = -7^\circ\,\mathrm{K}.$$

Thus, $\Theta_p < 0$ for the polycrystalline sample and for the single crystal when the magnetic field direction is perpendicular to the c-axis, indicating the possibility of the existence of the antiferromagnetic interaction in this compound.

3. Thus, the results of an investigation of the temperature dependence of the magnetic susceptibility of praseodymium diselenide showed that its magnetic properties were governed primarily by localized 4f-electrons. A strong anisotropy of the magnetic susceptibility and of the paramagnetic Curie point were also established and it was found that the Curie point was negative for the polycrystalline sample and for the single crystal along the direction perpendicular to the c-axis.

In conclusion, the authors express their gratitude to Professor E. I. Kondorskiĭ for discussing the results of the present investigation and for his valuable comments, and to A. A. Eliseev for carrying out the x-ray measurements.

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