THE PELTIER- AND SEEBECK-EFFECT OF $Rb_{0.3}MoO_3$ IN THE DEPINNED CHARGE-DENSITY-WAVE STATE

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ABSTRACT

The four thermoelectric Onsager-coefficients have been measured in the sliding charge-density-wave (CDW) state of $Rb_{0.3}MoO_3$. All coefficients except the one related to the heat conductivity display a strong electric field dependence above the same threshold field. The off-diagonal coefficients violate Onsager's symmetry relation in the nonlinear region. The results are discussed in an interacting two-fluid model with the heat transport by the sliding CDW neglected.

The measurements[1-3] of the electric field dependence of the Seebeck-effect have been motivated by an expected analogy with superconductors where the condensed electrons carry charge but no entropy. Surprisingly, these measurements yielded a strongly electric field dependent thermoelectric Onsager-coefficient indicating a per electron CDW heat transport comparable to or larger than the Peierls-gap. This finding suggests a boil-off of the CDW condensate in conflict with the X-ray diffraction result[4] of a field independent CDW order parameter.

The purpose of the present study is to resolve the above discrepancy by measuring in a $Rb_{0.3}MoO_3$ crystal the electric field dependence of all the four Onsager-coefficients, L_{ij}, appearing in the thermoelectric transport equations

$$j = L_{11}(E)\,\frac{E}{T} - L_{12}(E)\,\frac{\nabla T}{T^2} \quad \text{and} \quad U = L_{21}(E)\,\frac{E}{T} - L_{22}(E)\,\frac{\nabla T}{T^2}\,, \tag{1}$$

where j and U are the electric and heat currents, E is the electric field, and T is the temperature. The measurements have been performed on a $Rb_{0.3}MoO_3$ single crystal in the temperature range of 60 to 80 K. The details of experimental techniques will be described elsewhere.

The electric conductivity, $\sigma = L_{11}/T$, displays a strong nonlinearity above a threshold field [Fig.1(a)]. The other diagonal coefficient, L_{22}, related to the heat conductivity, has been found to be field independent

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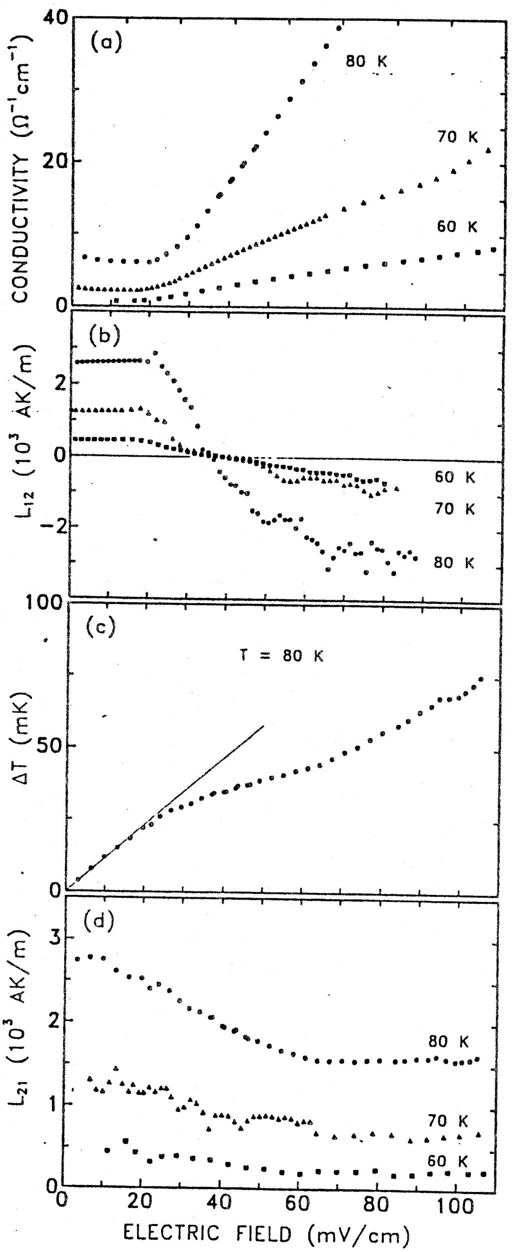

FIG.1 *The electric field dependence of conductivity (a), of L_{12} (b), of the temperature difference between sample ends (c), and of L_{21} (d).*

within an error bar of 5% in agreement with previous studies.[5]

Of primary interest for the present study are the off-diagonal coefficients L_{12} and L_{21}. The field dependence of L_{12}, related to the Seebeck-effect, is determined by subtracting the current-voltage (I-V) curves measured with and without a temperature gradient. The field dependence of L_{12} is shown in Fig. 1(b). L_{12} is strongly field dependent above E_T in accordance with Ref.3. It changes sign at $E \simeq 2E_T$ at all temperatures investigated.

To determine the field dependence of L_{21}, we have measured the temperature difference ΔT between sample ends due to the Peltier-heat generation and absorption at the contacts. ΔT *versus* E is shown in Fig.1(c). $L_{21}(E)$ is then calculated using $AL_{21}(E)\ E/T = K_{eff}\Delta T$ where K_{eff} is the field independent heat conductance of the sample and heat leaks. Like L_{12}, L_{21} starts to decrease above E_T [Fig.1(d)]. The changes of the two quantities, however, are not equal, i.e., the Onsager-relation $L_{12} = L_{21}$ is violated. Above about $2E_T$, even the signs are different. The violation of the Onsager-relation in a nonlinear system is not contrary to the theoretical expectations. The only thermodynamic constraint imposed on the coefficients, namely that the entropy production should be positive, is satisfied by the measured coefficients in the whole electric field range.

In the following we present an analysis in an *interacting* two-fluid model which does not conflict with the expectation of negligible CDW heat transport. Artemenko *et al.*[6] were the first to introduce such a model to interpret their nonlinear Hall-effect data in TaS_3. They pointed out that the CDW current induces a backflow of normal electrons so the total normal current is $j_n = \sigma_n E - \alpha j_{CDW}$, where σ_n is the low-field conductivity and α is a coupling constant. Here we extend this model by the assumptions described by the following equations:

$$j_n = \sigma_n E - L^n_{12}(\nabla T/T^2) - \alpha j_{CDW} \quad (2)$$

$$j_{CDW} = \sigma_{CDW} E - \beta j_n \quad (3)$$

$$j = j_n + j_{CDW} \quad (4)$$

$$U = L^n_{12}/(T\sigma_n)\, j_n \qquad (\nabla T = 0) \quad (5)$$

The normal electrons couple to the electric field and temperature gradient by the field-independent coefficients σ_n and L^n_{12}, respectively, and they also couple to the CDW current inducing a backflow of $-\alpha j_{CDW}$. The CDW couples only to the electric field and normal current but - in our approximation - not to the temperature gradient. The coupling term $-\beta j_n$ in (3) has not been used before, its existence is obviously a direct consequence of the similar term in (2). The heat current in Eq.(5) is only carried by the normal electrons in accordance with Eq.(3), i.e., $\Pi_{CDW} = 0$. Equation (5) utilizes that from the Onsager-relation valid in the low-field linear region $\Pi_n = L^n_{12}/(T\sigma_n)$.

σ_n and L^n_{12} are easily determined by low-field measurements. According to (3), the field dependence of the heat current [Fig.1(c)] reflects the field dependence of the normal current which is nonlinear due to the CDW - normal electron coupling. From the measured U-V and I-V curves α can be inferred. By comparing j as a function of E and ∇T to Eq.(1), the measured L_{12} coefficient is obtained as

$$L_{12} = \frac{1 - \beta}{1 - \alpha\beta} L^{n}_{12}.$$

In this picture the field dependence of L_{12} is the consequence of the field dependence of the couplings α and, more importantly, β. The magnitude of α is in good agreement with the Hall-effect result[7] of $\alpha \cong 0.1$ at $T = 80$ K.

In conclusion we have measured the electric field dependence of the four thermoelectric Onsager-coefficients of $Rb_{0.3}MoO_3$. L_{22} is found to be field independent in agreement with previous studies. Both L_{12} and L_{21} are strongly field dependent in the nonlinear conductivity region, the Onsager-relation, however does not hold, i.e., $L_{21} \neq L_{12}$ if $E > E_T$. We interpret our results in an interacting two-fluid model with two couplings between the normal and CDW currents. The heat transported by the sliding CDW is neglected in accordance with the theoretical expectation of $\Pi_{CDW} \ll \Delta$. The results are in good agreement with nonlinear Hall-effect measurements.

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