^{87}Rb NMR spin-lattice relaxation in the charge-density wave phase of $Rb_{0.3}MoO_3$

P. Matus, P. Bánki, and G. Kriza
Research Institute for Solid State Physics and Optics, P.O.B. 49, H-1525 Budapest, Hungary

Abstract. The ^{87}Rb NMR spin-lattice relaxation rate (SLRR) has been measured in the charge-density wave (CDW) phase of the quasi-one-dimensional conductor $Rb_{0.3}MoO_3$ in a magnetic field of 2 T perpendicular to the conducting chains. The SLRR exhibits a peak at the critical temperature T_c = 180 K, attributed to the critical fluctuations at the phase transition. Two more broad peaks are observed within the CDW phase: one peak at 150 K (0.8 T_c) and another one at 60 K (0.3 T_c). We discuss the 0.8-T_c peak in terms of quantum coherence effects in analogy with the Hebel-Slichter peak in superconductors. The 60 K peak is attributed to the phase excitations of the CDW.

NMR measurements in charge-density wave (CDW) systems have provided a wealth of information on the structure and excitations of the CDW [1]. In this article we address a so far largely unexplored problem: We ask what conclusion can be drawn on the collective- and quasiparticle excitations of the CDW from an analysis of the spin–lattice relaxation rate (SLRR). The system investigated is the so-called blue bronze $Rb_{0.3}MoO_3$, a quasi-one dimensional conductor. This is one of the most studied CDW systems [2], and the transition from a high-temperature metallic phase to a low temperature incommensurate CDW phase at T_c = 180 K is well documented and understood [2].

We have measured the ^{87}Rb NMR spectrum and SLRR of a $Rb_{0.3}MoO_3$ single crystal both in the metallic and CDW states. The nuclear spin of ^{87}Rb is I = 3/2, therefore the quadrupolar interaction with the electric field gradient (EFG) tensor in the monoclinic crystal has to be taken into account in analyzing the NMR properties. With respect to crystal symmetry, the 6 Rb nuclei in the unit cell of the crystal are situated in two inequivalent sites: Two nuclei are coordinated by 10 oxygen atoms, and the remaining four nuclei by 7 oxygen atoms giving rise to two sets of quadrupole-splitted lines. Each set consists of the $m = -1/2 \leftrightarrow +1/2$ "central transition" as well as two "satellites" $\pm 1/2 \leftrightarrow \pm 3/2$. All our measurements have been performed on the central transition of the 7-fold coordinated nucleus. The static magnetic field B_0 = 1.94 T has been applied in the a-c plane (perpendicular to the conducting b direction) at an angle of 10° from the $[\bar{2}01]$ direction (see the inset of Fig. 1), which is one of the principal axes of the room-temperature shift tensor [3].

Two typical spectra—one taken above and one below T_c—are shown in Fig. 1. The narrow line of the metallic state broadens in the CDW phase to a "double-horn" spectrum characteristic of an incommensurate structural modulation [1]. The primary reason of the broadening is that the periodic lattice distortion accompanying the CDW modulates the components of the EFG tensor.

The SLRR has been measured with the saturation recovery technique. The recovery of the magnetization M as a function of time t is strongly non-exponential. To infer an average relaxation time T_1, we have fitted a stretched exponential form $\Delta M = M_0 \exp[-(t/T_1)^{\beta}]$ to the recovery curves with the limits of the fit chosen self-consistently as $0.1\, T_1 < t < 10\, T_1$. The SLRR relative to the temperature, $1/(T_1 T)$, obtained in this procedure is shown in Fig. 2 as a function of temperature T. The SLRR exhibits three marked features: There is a sharp peak at the CDW transition, and there are two more broad peaks *within* the CDW phase at 150 K and at 60 K.

In the metallic phase, the SLRR dominantly originates from the electronic quasiparticle excitations via the Korringa mechanism. The appearance of a peak at T_c is natural since the divergent charge fluctuations are coupled to a soft phonon mode, which, in turn, is coupled to the nuclei via the quadrupolar interaction. In the CDW phase, both the quasiparticle and collective mode excitations are expected to contribute to the SLRR. The number of quasiparticle excitations is exponentially small if $T \ll T_c$, but in the vicinity of T_c the quasiparticle contribution is comparable to the SLRR in the metallic phase and thus cannot be neglected. The quasiparticle contribution in the CDW state reflects the coherent nature of the CDW ground state which is manifested in a so-called Type II coherence factor in the expression for the SLRR [2]:

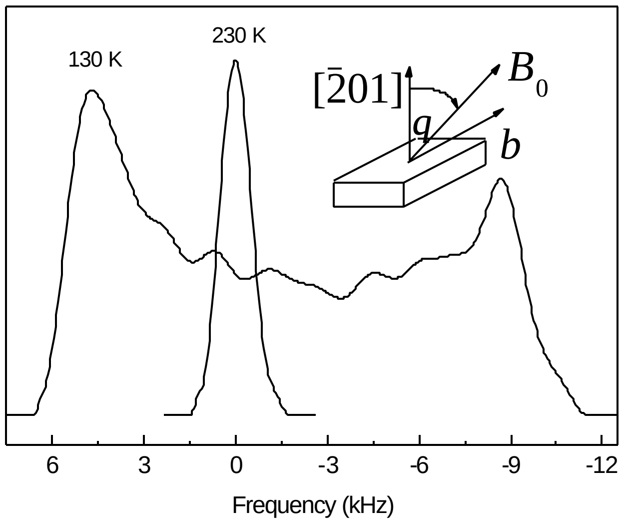

Figure 1

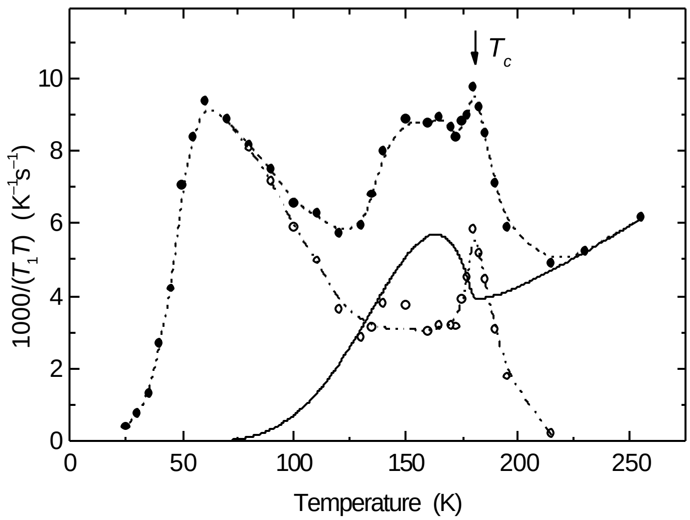

Figure 2

Figure 1: NMR absorption as a function of frequency relative to the carrier n_0 = 27.076 MHz above (230 K) and below (130 K) the temperature of the CDW transition. (The relative magnitudes of the two spectra are scaled for the clarity of the figure.)

Figure 2: Temperature dependence of the spin–lattice relaxation rate (full circles). The temperature of the CDW transition is indicated by an arrow. In the CDW phase, the SLRR is divided into two parts. The solid line displaying a coherence peak is the quasiparticle contribution computed using Eq. (1). The open circles represent the collective mode contribution. The dotted lines are guides to the eye.

$$\frac{1}{T_1 T} = \frac{1}{T_1 T}\bigg|_n \frac{2}{T} \int_\Delta^\infty \frac{E\,dE}{\sqrt{E^2 - \Delta^2}} \int_\Delta^\infty \frac{E' dE'}{\sqrt{E'^2 - \Delta^2}} \left(1 + \frac{\Delta^2}{EE'}\right) \frac{f(E)[1 - f(E')]\Gamma}{(w_L + E - E')^2 + \Gamma^2} \ , \qquad (1)$$

where $1/(T_1T)|_n$ is the SLRR in the normal (metallic) phase, Δ is the CDW gap, f is the Fermi function, w_L is the Larmor frequency, and Γ is the damping parameter. The details of the temperature dependence obtained from Eq. (1) depend on the temperature dependence of Δ and Γ as well as the energy dependence of Γ, but a general feature is a peak—called Hebel–Slichter peak in the context of superconductors—at about 0.8…0.9 T_c. This is illustrated by the solid line in Fig. 2, computed using Eq. (1) with $\Delta(T) = \Delta_0 \sqrt{1 - T/T_c}$, Δ_0 = 700 K, and Γ = 70 K.

The collective mode contribution is estimated by subtracting the calculated quasiparticle contribution from the measured SLRR. The curve obtained this way (open circles in Fig. 2) resembles strongly the SLRR of spin density wave systems, where the dominant contribution has been shown to originate from the phase excitations of the density wave [4]. This assignment is in agreement with our observation that T_1 is longer at the edges of the spectrum than in the middle. In this framework, the 60-K peak occurs because at this temperature the average phason relaxation time is equal to the inverse Larmor frequency [5].

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References

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